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Application of Nuclear Radiation to Industry*

By JOHN R. MENKE

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NUCLEAR PARTICLES as tools for experimental investigations are not new. Hevesy (*1*) studied the chemistry and solid-state physics of lead, beginning in 1912, by using the emanations from radioactive lead, which is found in nature as a disintegration product of U, Th, and Ac. Furthermore, artificial radioactive materials (*2*) were known in the laboratories for many years before World War II. The new feature in the field is the cheapness and convenience, the variety and the strength of the materials which are available today from nuclear reactors (*3*).

It is this greatly increased availability of new tools that presents a significant opportunity to industry. It is, however, only a partially grasped opportunity. For instance, out of some 5,000 isotope shipments in the past two and a half years by the U. S. Atomic Energy Commission (*4*) about 3.4% (about 170) were for industrial use (if we exclude agriculture and other biological purposes). Conditions contributing to this situation include a scarcity of trained personnel, and some sensitivity to the unfamiliar safety and publication requirements. A free and unclassified school has been organized and run by the Oak Ridge Institute of Nuclear Studies to help improve the situation with respect to qualified personnel.

It may be that a fuller exposition of the potentialities of these new tools can serve a useful purpose here. We shall look into these potentialities by following a typical life-history of a nuclear particle and by examining the functions (*5*) that it might perform at each step. In addition, we shall look at an example or two of each type of function. In this manner we may hope to find the seeds of new applications, of solutions to particular problems which might be missed by a simple recitation of existing or proposed applications. Several good lists already exist in the literature (*4*, *6*, *7*, *17*, *18*, *23*).

By definition, a nuclear particle is born in a nuclear event, in some rearrangement of the nucleus of the atom which expels the particle. The particle then generally travels through matter, affecting the matter and, in turn, being affected by it. It may lose its identity in the medium or escape from the region of interest. Or, more rarely, it may combine and react with another nucleus.

All the potential functions of the particle and a useful outline for us are contained in this simple cycle. The type of particle and the manner in which it is expelled is usually characteristic of the source nucleus and can indicate the identity of the source. (For instance, an alpha particle is emitted with an energy of 5.3 Mev and half-life of 136 days when polonium decays to lead.) The particle will affect matter by transferring part of its energy, *i.e.*, by ionizing or displacing atoms and molecules in or near its path. (Thus, this 5.3-Mev alpha particle will create a few times 10^4 ion pairs per cm in air.) And the particle itself will be affected by the matter through which it can penetrate, losing its energy, being scattered and perhaps absorbed. (The alpha particle will be stopped in

* From a talk presented at the annual meeting of the Institute of Radio Engineers.

about 4 cm in traveling through normal air.) Stopped and deionized it loses its identity as a nuclear particle; then, as a helium atom, it adds to our source of helium. It may, however, react directly with another nucleus to form a somewhat different combination, usually so unstable as to cause a new cycle of such events. [The alpha reaction, $N^{14}(\alpha,p)O^{17}$, was the first nuclear reaction discovered.]

An outline of functions then will have four fundamental categories, *i.e.*, uses dependent upon:

1. Identifying characteristics of a nuclear event (such as indication of source)

2. Effects of nuclear particles on matter (such as ionization or displacement of atoms and molecules in medium)

3. Effects of matter on nuclear particles (such as penetration through, and scattering and absorption by, medium)

4. Reactions of nuclear particles with nuclei (such as the transmutation of elements)

All except the fourth group have seen some important industrial applications. Perhaps the greatest number of different types of new applications fall into the first class; the second and third classes have long been served by natural radioactive sources.

Identifying characteristics of a nuclear event. Natural radioactivity has long been used to identify its source. Thus we have the familiar field identification of uranium and thorium for the prospector. More recently the naturally radioactive potassium (K^{40}) has been used in assaying its salts. These techniques can be made semiquantitative in the field and can be made rigorously quantitative in the laboratory.

$_{81}$Tl and all the heavy elements above it in the periodic table ($_{82}$Pb, $_{83}$Bi, $_{84}$Po, $_{85}$At, $_{86}$Rn, $_{87}$Fr, $_{88}$Ra, $_{89}$Ac, $_{90}$Th, $_{91}$Pa, $_{92}$U) have natural radioactive isotopes. At least five lighter elements are also known in naturally radioactive forms, $_{19}K^{40}$, $_{37}Rb^{87}$, $_{62}Sm^{152}$, $_{71}Lu^{176}$ and $_{75}Re^{187}$. Of these seventeen elements, only lead, bismuth and potassium, are of major interest to industry. These were all that were available before the advent of artificial radioactivity and constituted a fundamental limit to applications of radioactivity. With them, however, the basic identification techniques were developed. A classic example of this type of work, pointed out by Stanley (*8*), is the experiment by Hevesy and Zechmeister in 1920 (*1*) demonstrating Arrhenius' theory of electrolytic dissociation. All lead isotopes, of course, exhibit nearly identical chemical properties. Subsequent separation of the two salts, both containing radiolead, proved the exchange of lead ions in solution. Hevesy also employed radiolead to study self-diffusion phenomena; there is probably no better technique today.

Artificial radioactivity, before the nuclear reactor, removed the previous limitations of variety but was an excessively expensive tool, being manufactured mostly by cyclotrons and with photoneutron sources at very low efficiencies.

Artificially radioactive sources from piles are cheap and include elements from a large fraction of the periodic table. The metallurgical, chemical, and geological industries (*4, 7, 8, 9, 13*) have been among the more active developers of the field. In steelmaking (*10, 11, 19*), for instance, the rapid and continuous determination and tracing of sulfur and phosphorus, the kinetics of slag-metal interfaces, the thermodynamic activities and diffusion phenomena in solids and liquids are all being examined with tracer techniques. In the rubber and oil industries the kinetics of complex organic reactions are being studied—especially vulcanization and its relation to sulfur and the catalytic

problems associated with the Fischer-Tropsch process. Phenomena of wear (*20*) and lubrication are being studied by the oil industry along with active work in logging oil wells and tracing oil routes by radioactivity.

Some more miscellaneous applications of the "characteristics" of the nuclear event include the use of the definite decay period as a measure of time. The otherwise inconvenient magnitude, 2×10^9 years—the so-called age of the earth—is measured by evaluation of the Rb-Sr and U-Pb ratios together with the known half-life of the radioactive fraction. An ingenious timing circuit (*5*) compensating for the decay of a primary radio source by balancing against the decay of a similar source has been described. Standards of energy for neutrons from photosources are also emerging. These standards make use of one of the remaining characteristic quantities in the radioactive event, the particle energy.

Over 80% of the new industrial uses of pile-generated radioactivity reported by the AEC (*4*) fell into this large indicating, tracing and use-of-the-characteristics-of-radiation category. It is a category whose potentialities defy presentation here; we could devote considerable space to it without giving the field adequate attention.

Effects of nuclear particles on matter. Nuclear particles affect the atomic and molecular structure of matter through which they may pass. One such effect is the ionization that the charged particles induce. It is the most usual means for their detection and measurement. This ionization is regularly put to use in the paper, textile and grain industries for the elimination of static electricity by means of alpha particles from radium and polonium. The static charge is a production nuisance in the manufacture of paper and textiles and is a danger of real magnitude in the handling of dry grain. This use is an interesting example, being one case where the older, natural radioactivity remains more potent and economic than the artificial activity. Ingenuity may yet change this relationship. The ionizing capability of particles can also be used to reduce the sparking potential of high voltage gaps and to provide cold electronic emission in special tubes. Furthermore, there is some possibility that ionizing radiation can be used to sterilize and even to produce mutations of value in biologic material. (Improved strains of various moulds have already been developed from radiation-induced mutants.)

Other effects can also be produced in matter—for instance, heating, excitation, and other energy release (*21*, *22*), coloring, and even displacement of atoms from their normal matrices. Energy uses, such as radioactive batteries, *etc.*, deserve an important place in this category. The ultimate chemical, physical and engineering utility of these effects or of what they might catalyze has not been well measured.

Effects of matter on nuclear particles. The first observed effect of nuclear particles was their penetrating ability. The gamma radiation from radium is a familiar tool in industrial radiography. At $20.00 per millicurie it is meeting competition from cobalt-60 with similar gamma radiation at about one tenth the cost. Since radiation emerging from matter carries information gathered along its path, such information can be used to detect, measure and control the character of the path. Devices have been developed which use beta particles to measure and control the thickness of moving sheets such as paper and plastic films. Similar thickness-measuring applications of other radiations are also known.

The information gathered by the effect of matter on radiation need not be due to absorption from a beam penetrating through the material. The

scattered component has been used with success where only one side was available as for instance in liquid level measurement. A suggestion has been made to locate buried survey stakes with a small amount of penetrating radiation and similarly to locate pipe scouring apparatus stuck in buried sections of long pipes. These examples only serve to illustrate some of the many such applications recently made feasible by new, cheap radiation sources.

Reactions of nuclear particles with nuclei. Finally, nuclear particles can be used to cause nuclear reactions. It is this capacity that has captured our imaginations—and rightly so. However, we should pause a moment short of the nuclear *chain* reaction to look at less spectacular reactions.

Brown and Goldberg (*14*) have used neutron sources to activate nuclei, transmuting them into radioactive materials that can frequently be readily detected and identified. The power of this analytic tool in combination with quantitative chemical analysis is shown by their results. Trace quantities of the order of a few parts per million have been measured with a standard deviation of five to ten percent. In some cases problems which have been unsolvable with spectrographic techniques can be neatly handled with ordinary pile neutron sources. A similar use of an artificial neutron (gamma-neutron) and (alpha-neutron) source has seen service in the search for oil. The source is lowered into a drill hole to remotely activate and analyze different strata.

It may be useful at this point to review the particles, indicating those significant in each category. (Table on right.)

Neutrinos are an interesting case of being altogether too good—they penetrate matter so well that they can neither affect nor be affected by reasonable masses interposed in their paths. The earth itself is probably just thick enough to stop some of them. Alpha particles are the most practical ionizers and neutrons are the best instigators of nuclear reactions. Alphas, betas and gammas are ordinarily most useful in identification procedures.

It is of interest to examine the special case of the nuclear reaction, the self-supporting neutron chain reaction which is the primary mechanism of the atomic bomb and of the controlled nuclear reactor. It is probably not useful to repeat here what has been described so well in the Smyth report (*15*) and has been restated too many times. It is enough to say that the pile neutrons are the most efficient source of many of the artificial radioactive materials whose uses have been discussed above, that the pile may be used as a neutron source to apply directly to materials of interest (as, for instance, in the Brown and Goldberg analytical methods described above), that the fission reaction is a source of energy (heat), of radiation (both a hazard and a potentially useful product as in sterilizing, *etc.*, above) and of the crude radioactive fission fragments which may again be used as particle sources. One significant concept, however, has been added to the public forum since the issue of the Smyth report. It is the so-called "breeding" mechanism and is worth extra attention.

The isotope uranium-235, 0.7% of

Uses of Nuclear Radiations

	Major	*Minor*
Characteristics, identification, *etc.*	α, β, γ	p, n, fission fragments
Ionization, displacement, *etc.*	α	β, γ, n, fission fragments
Penetration, scattering	β, γ, n, neutrinos	α
Nuclear reaction activators	n	α, γ

natural uranium, is the only naturally occurring substance that can be fissioned with most of the neutrons available within a pile. If it were the only possibility, it would constitute a severe limitation. However, neutrons can transmute the source materials, thorium-232 and uranium-238 into the fissionable materials uranium-233 and plutonium-239. If enough neutrons can be made available, the much more abundant source materials can be rendered as fissionable, and as useful as the scarce isotope, uranium-235. Thus, the neutron economy of the reactor determines, to an important extent, the utility of a great resource.

The neutron balance in a reactor can be illustrated as follows: *Neutrons needed per fission*—(*a*) one neutron is needed to cause a fission; (*b*) one neutron at least is needed to replenish the fissioned atom in order to replace it and "break even" on fissionable material; and (*c*) on the average a fraction of a neutron is wasted in parasitic absorption and in escape from the pile. We can call the total needed $2 + x$ where x is a fraction less than unity. The neutrons produced per fission event we shall call $2 + y$ where y is also a fraction less than unity. If the amount produced per fission, $2 + y$, is greater than the amount needed to break even, $2 + x$, we can increase the total amount of fissionable material available, producing more fissionable material than we burn (at the cost of a little source material). There is more than a faint hope that breeding can be accomplished. This is one of the central technical problems (*16*) of large scale, economic atomic energy.

Finally, it should be emphasized that this report surveys the present status of an art which is in vigorous development. It is, therefore, a limited report which will, one expects, need much revision in the next few years.

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Comparative Survey of Ion Guns—I

Design of ion guns is among the more important problems in nuclear physics. In accelerating devices, as well as in mass spectrography, strong, easily focused beams are needed. This paper, which is the foundation for the two papers to follow it, discusses fundamentals of good ion-gun design

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THE ION GUN delivers a flow of ions to be (1) accelerated in a device such as a cyclotron, high-voltage tube, cavity accelerator, *etc.*, or, (2) analyzed by a mass spectrograph.

In the first case, only a few kinds of ions are to be produced: (a) protons or hydrogen atomic ions (p or $_1H^1$), (b) deutons or deuterons or heavy hydrogen atomic ions (d or $_1H^2$), and (c) helions or alpha particles or twice-charged helium ions (α or $_2He^4$) and sometimes lithium ions. In some cases, ions of tritium (mass three hydrogen) or light helium are used, but these are generally obtained by nuclear processes.

In the second case, every kind of ion may be wanted.

In every case where it is possible, the atoms to be ionized are brought into the ion gun in the form of a gas flow. If not, a solid cup is coated with the material to be ionized. We make no attempt to review this problem, and suppose that it is always possible to deliver the material to the source in the desired form.

In the gun, electrons are emitted by a cathode, which may be cold or hot, coated or not, and are accelerated through an electric field toward the atoms that are to be ionized. In the case of gaseous material, an electrical discharge is frequently used to increase the rate of ionization.

Mean Free Path—The problem of the mean free path for ionization is fundamental. Let us call it λ_i, and d the mean distance from the cathode to the walls. The probability of ionization is, for a single electron emitted by the cathode, approximately d/λ_i, when d is inferior to λ_i. If the pressure in the ion source were equal to that of the accelerating device (λ_i equal to several hundred meters), the rate of ionization would be exceedingly low. Let us take d equal, for instance, to 10 cm and $\lambda_i = 200$ m; then, we will create approximately one ion for 2000 electrons, and, as the rate of removal is generally low, we will get very few useful ions.

In other words, although the pressure in the accelerating device or the mass spectrograph must be sufficiently low to allow the ions to travel from the source to the target without colliding with neutral atoms, the electrons emitted by the cathode must have a high probability of colliding with neutral atoms to give a high rate of ionization. Thus, one may (a) introduce a sufficiently high difference of pressure between the source and the high vacuum, (b) force the electrons to

describe very complicated paths before reaching the walls, or (c) have a combination of these two principles. In cases (a) and (c), a continuous flow of gas must pass from the ion source to the high vacuum. High-speed pumping is, therefore, necessary.

Gas Flow—The problem of the gas flow may be studied by Knudsen's formula. It states that

$$Q = \frac{\pi}{3\sqrt{2}} \frac{D^3}{L} \sqrt{\rho_1} (P_A - P_B) \quad (1)$$

where Q is the gas flow in kg/sec,
D is the hole diameter in m,
L is the hole length in m,
ρ_1 is the specific mass of gas at unit pressure [kg/m³:kg/(m × sec²)]
($\sqrt{\rho_1}$ has the dimensions of the reciprocal of a speed)
P_A, P_B are the pressures at both ends of the hole [kg/(m × sec²)]

Knudsen's formula gives the flow of gas when the mean free path is superior to the diameter of the hole. In the case of ion sources, P_B is generally 0.1 to 1.0% of P_A; therefore we may neglect it.

For calculating the pumping speed, it is interesting to have the velocity of flow V in m³/sec at the pressure P_B

$$V = \frac{Q}{\rho_B} = \frac{Q}{\rho_1 P_B}$$

$$= \frac{\pi}{3\sqrt{2}} \frac{D^3}{L} \frac{1}{\sqrt{\rho_1}} \frac{P_A}{P_B} \quad (2)$$

Practically, the flow from the ion source is approximately 50% of the total flow (taking into account outgassing and leaks in the high vacuum chamber).

We may obtain another formula by calculating ρ_1. We have $\rho_1 = 1/rT$ and $r = \mathfrak{R}/\mathfrak{M}$, where $\mathfrak{R}$ is the constant of gases for one molecule (8,320 J/°K), and $\mathfrak{M}$ the molecular mass. Thus

$$V = \frac{\pi}{3\sqrt{2}} \frac{D^3}{L} \frac{P_A}{P_B} \sqrt{\frac{\mathfrak{R}T}{\mathfrak{M}}} \quad (3)$$

In most cases, we may put $T = 300$°K; then Eq. 1 and Eq. 3 become

$$Q = 2.25 \frac{D^3}{L} P_A \sqrt{\mathfrak{M}} \quad (4)$$

$$V = 1.17 \frac{D^3}{L} \frac{P_A}{P_B} \frac{1}{\sqrt{\mathfrak{M}}} \quad (5)$$

where D and L are in mm, P_A in 10^{-2} mm Hg, Q in mg/hour and V in liters per sec. For deuterium ($\mathfrak{M} = 4$), for example, if $D = 1$ mm, $L = 8$ mm, $P_A = 2 \times 10^{-2}$ mm Hg, $P_B = 10^{-6}$ mm Hg, then $V = 146$ liters per sec. The flow for such a hole is very important, and high-speed pumping devices must be provided. With the same values of the parameters, the flow in mass is 1.12 mg/h.

This is rather a low consumption of deuterium; but its price is not at all negligible. The true consumption is always higher because of losses in the apparatus. Let us now compare the flow of gas with that of ions. Assuming 1 ma (a rather high value) and taking into account the specific charge of deuterons 4.77×10^7 coul/kg, we get a mass of 0.2×10^{-10} kg/sec, *i.e.*, 6.5% of the mass of neutral gas. The true rate is generally lower.

EDITOR'S NOTE: *The three papers in this series combine a description and comparison of some kinds of ion guns which have been described in the literature since Thomson's experiments in 1913. In the first paper, appearing here, the general properties and specific characteristics of typical guns are discussed. In the two papers appearing in following months, the authors describe the properties of representative ion sources, show the merits of each type, and review the necessary refinements of ion-gun design.*

Requirements

In addition to these general features of ion sources, there are various specific requirements that depend on the particular application of the source.

Purity of Beam—In the case of mass spectrography, it is generally interesting for the source to deliver many kinds of ions of the gas which is studied; sometimes, mercury ions or hydrocarbon ions (from the pump oil) are also desired for comparison (doublets).

But, when the source is connected to an accelerating device, this is not the case; only one kind of ion is wanted. For example, if we want to accelerate protons, molecular ions of hydrogen are undesirable because their mass is twice that of protons and they reach only 70% of their final speed. The result is that the rate of nuclear transmutations is considerably decreased below that for pure proton beams. Or, if we accelerate helions, single helium ions (He^+) are undesirable because their final energy is only half of that of helions for the same accelerating potential. Ion guns frequently give high rates of those undesirable ions.

Introducing some kinds of impurities that ionize at low voltage, such as mercury vapor, gives a manifold increase of the beam current. Unfortunately, magnetic analysis shows that almost all the ions arise from these impurities, so that the useful current is actually decreased. Several investigators pointed out that water vapor in hydrogen gives an increase of the proton rate.

Other Features—Among the other characteristics of an ion gun is its electrical power consumption. It must be low, principally in the case of accelerating devices, because the target must always be grounded; hence the source is at high potential, and power must be delivered through insulation transformers. The control of the source must, for the same reason, be as simple as possible, because it must be made from the observation chamber, several meters distant from the source.

Great stability is also required, principally in the cases of precision measurements and accelerating devices (measurements of energy levels) as in mass spectrographs (measurements of relative abundances). It is obvious that the accuracy of such measurements depends upon the stability of the source.

Speed of Ions—If only small potentials are used in the source (that is, small by comparison with the voltages applied in further devices), the speed of removed ions is negligible and one may have monokinetic beams. Electrostatic Van de Graaff generators give high potentials that are steady within 0.01% if they are provided with adequate control devices. If the ions are monokinetic one can explore resonance rays with great accuracy. Some mass spectrographs give only focusing of directions, not of speeds. It is desirable that the ion speeds be practically equal for good concentration on the measurement apparatus.

Cathode—Low-voltage sources generally require hot cathodes, which are a source of much trouble to experimenters. The mean life of the best cathodes is always low, because, at normal rates, the electronic emission is not sufficient to protect the cathode fully from ionic bombardment. The cathode problem is the most important one for all types of low-voltage sources. It is always necessary to provide a device that permits replacing dead cathodes. On the other hand, high-voltage sources can use cold cathodes, which give very much longer life.

Summary of Requirements—A good ion source should yield a large ion current, allow focusing in a narrow beam, give a high rate of some desired type of ions, have a low gas consumption, require low electrical power consumption, and give monokinetic ions.

Introduction to the Theory of Diffusion and Slowing Down of Neutrons—I*

By R. E. MARSHAK,† H. BROOKS,‡ and H. HURWITZ, Jr.‡

To design a chain-reacting system properly, one must be able to predict the behavior of the neutrons in the assembly. In particular, one must calculate the size of unit necessary to sustain a chain reaction, the distribution of fission throughout the assembly, and the effect on reactivity of various changes which may occur in the assembly in the course of operation.

The methods of performing these calculations vary considerably with the nature of the nuclear reactor, and the techniques are in some cases rather specialized. All the methods, however, have the common feature of tracing the history of the neutrons from the time they are produced in a fission process until they either cause another fission, are captured, or escape from the pile. Two of the most important tools for calculating the neutron behavior are *diffusion theory* (applicable when the neutrons may be considered to be monoenergetic) and *age theory* (applicable when the neutrons slow down through elastic collisions with nuclei).

The fundamental equations of diffusion theory and age theory are obtained by making certain approximations which are justified in many, but not all, of the problems that one encounters in reactor design. When the conditions for the validity of diffusion theory and age theory are not fulfilled, one must use some of the more sophisticated lines of attack which are available.

The purpose of this series of papers is to present a systematic development of diffusion and age theory, starting with the Boltzmann equation. An attempt is made to underline the approximations used in deriving these theories, and thereby to specify the limits of their validity. The subject of how corrections can be made to these theories whenever necessary and how more accurate theories can be formulated and applied, is also discussed. Parts I and II of this series of papers will cover diffusion theory; Parts III and IV will cover age theory.

Much of the material presented is the result of research in this country and abroad during the war years. Groups under the direction of Peierls in England, Wigner and Fermi at Chicago, Placzek at Montreal, Serber at Los Alamos, and Nordheim at Oak Ridge have made important contributions to the subject. In many cases the development of particular techniques occurred almost simultaneously in several laboratories. The authors have made no attempt to provide a complete list of references but have limited themselves to indicating a few of the articles where further information may be found.

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* This is the first of a series of four papers compiled from notes taken by H. Brooks and H. Hurwitz, Jr., on lectures given at the General Electric Company by R. E. Marshak during the summer of 1946. A large part of the work described was performed under the auspices of the Atomic Energy Project and thus will be presented in volumes of the National Nuclear Energy Series (Manhatten Project Technical Section).

Parts II, III, and IV will be published in NUCLEONICS in succeeding months.

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The Diffusion Theory

In deriving the diffusion equation, we start with the Boltzmann equation, which is an accounting between the number of particles leaving and entering a given space and velocity range, more strictly a given infinitesimal volume of phase space. We suppose that all collisions are perfectly elastic and involve no change in energy. Let the number of particles found in a volume element $d\mathbf{r} = dxdydz$ at $\mathbf{r}$, with their velocity vector directed within a solid angle $d\mathbf{\Omega}$ at $\mathbf{\Omega}$, be denoted by:

$$N(\mathbf{r},\mathbf{\Omega},t)d\mathbf{r}d\mathbf{\Omega}$$

Then form 1 of Boltzmann's equation is:

$$\frac{DN}{Dt} = -NV\sigma + \int N(\mathbf{r},\mathbf{\Omega}',t)V\sigma_s F(\mathbf{\Omega}\cdot\mathbf{\Omega}')d\mathbf{\Omega}' + S(\mathbf{r},\mathbf{\Omega},t) \quad (1)$$

The term on the left-hand side of Eq. 1 is the change in the number of particles in a volume element which moves with the particles going in the direction $\mathbf{\Omega}$. It is related to the local change in particle density by the usual hydrodynamic relation:

$$\frac{DN}{Dt} = \frac{\partial N(\mathbf{r},\mathbf{\Omega},t)}{\partial t} + V\mathbf{\Omega}\cdot\nabla N(\mathbf{r},\mathbf{\Omega},t) \quad (2)$$

The second term on the right-hand side of Eq. 2 thus represents the convection of particle density into a given volume element. The first term on the right-hand side of Eq. 1 is the total number of particles removed from the volume element of phase space by scattering and capture, σ representing the total cross section per cm³ of material. The second term is the number of particles scattered into the given volume element of phase space from other velocity directions. The function $F(\mathbf{\Omega}\cdot\mathbf{\Omega}')$ represents the relative probability of scattering through an angle whose cosine is $\mathbf{\Omega}\cdot\mathbf{\Omega}'$, where $\mathbf{\Omega}'$ is a unit vector in the direction of the initial velocity and $\mathbf{\Omega}$ is a unit vector in the direction of the final velocity. The function F is normalized in such a way that its integral over all possible initial or final directions is unity, that is:

$$\int F(\mathbf{\Omega}\cdot\mathbf{\Omega}')d\mathbf{\Omega}' = \int F(\mathbf{\Omega}\cdot\mathbf{\Omega}')d\mathbf{\Omega} = 1 \quad (3)$$

The last term on the right-hand side of Eq. 1 is a source term denoting the number of neutrons originating in the given element of phase space per second. The quantity σ_s is the cross section for scattering alone. Fission is neglected because it does not introduce any essentially new features into the problem.

It is illuminating to obtain steady-state solutions of Eq. 1 when one has plane symmetry for sources with an isotropic velocity distribution. The steady-state assumption corresponds to setting $\partial N/\partial t = 0$; the plane symmetry and the isotropic source assumptions correspond to:

$$S(\mathbf{r},\mathbf{\Omega},t) = S(z);$$
$$N(\mathbf{r},\mathbf{\Omega},t)d\mathbf{r}d\mathbf{\Omega} = N(z,\mu)dzd\mu d\phi \quad (4)$$

where μ is the cosine of the angle of the velocity vector with the z direction, and ϕ is really the longitude of the velocity vector as shown in Fig. 1.

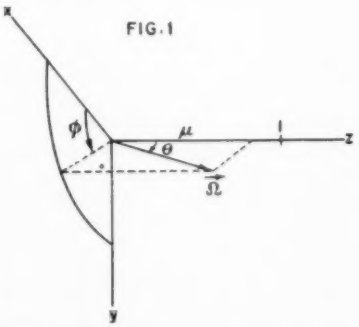


FIG. 1

We define the neutron density $N_0(\mathbf{r},t)$ as follows:

$$N_0(\mathbf{r},t) = \int N(\mathbf{r},\mathbf{\Omega},t)d\mathbf{\Omega} \quad (5)$$

or, for the particular assumptions, above:

$$N_0(z) = 2\pi \int_{-1}^{+1} N(z,\mu)d\mu \quad (6)$$

With our assumptions the Boltzmann equation takes the form:

$$V\mu \frac{\partial N(z,\mu)}{\partial z} = -N(z,\mu)V\sigma + \int N(z,\mu')V\sigma F(\mu_0)d\Omega + S(z) \quad (7)$$

where μ_0 is the cosine of the angle between the initial and final velocities and is found from the trigonometric relation:

$$\cos\theta_0 = \cos\theta\cos\theta' + \sin\theta\sin\theta'\cos(\phi - \phi') \quad (8)$$

or

$$\mu_0 = \mu\mu' + \sqrt{1-\mu^2}\sqrt{1-\mu'^2}\cos(\phi - \phi')$$

Solution of Boltzmann's Equation

Diffusion theory approximation. We suppose that the collision function $F(\mu_0)$ may be expanded in a series of spherical harmonics:

$$F(\mu_0) = \sum_{l=0}^{\infty} \frac{2l+1}{4\pi} F_l P_l(\mu_0) \quad (9)$$

with $F_l = \int F(\mu_0)P_l(\mu_0)d\Omega$. A similar expansion is possible for the phase density function:

$$N(z,\mu) = \sum_{l=0}^{\infty} \frac{2l+1}{4\pi} N_l(z)P_l(\mu) \quad (10)$$

with $N_l(z) = \int N(z,\mu)P_l(\mu)d\Omega$.

We now assume that the function $N(z,\mu)$ is almost isotropic; we shall see that this will be the case provided the following conditions are fulfilled:

1. We are far from a source, *i.e.*, far compared to a mean free path.

2. We are far from any boundaries, *i.e.*, discontinuities in the scattering medium, or, more precisely, regions in which the properties of the medium change appreciably within a distance comparable to a mean free path.

3. Probability of capture is small compared to probability of scattering.

It is not obvious that the near isotropy of $N(z,\mu)$ follows from the above assumptions. What we shall actually do is to solve Boltzmann's equation *assuming* that N is nearly isotropic and later show that this solution closely approximates the rigorous solution if the above three conditions are fulfilled.

It is clear that $N(z,\mu)$ cannot be completely isotropic, that is, independent of μ, for if the particles were moving with equal probability in all directions, there could be no net flow and hence no diffusion. What we do is to assume the minimum possible anisotropy in N which still yields a net particle current. This corresponds to:

$$N(z,\mu) \cong \frac{1}{4\pi}[N_0(z) + 3\mu N_1(z)] \quad (11)$$

with $N_0(z) = \int N(z,\mu)d\Omega$ = particle density and $N_1(z) = \int \mu N(z,\mu)d\Omega = J/V$, where J = particle flux or current.

For economy in writing we choose our units so that $V = 1$ and $\sigma = 1$, and define

$$1 - f = \frac{\sigma_s}{\sigma} \quad (12)$$

The Boltzmann equation then takes the form (form 2):

$$\mu\frac{\partial N}{\partial z} = -N + (1-f)\int N(z,\mu')F(\mu_0)d\Omega' + S(z) \quad (13)$$

Let us integrate this equation over $d\Omega$:

$$\frac{dN_1}{dz} = -N_0 + (1-f)N_0 + S_0(z) \quad (14)$$

$$S_0(z) = 4\pi S(z)$$

The origin of all the terms in Eq. 14 is obvious except the second term on the right-hand side. To see this we must use the expansions in Eqs. 9 and 10. In Appendix A the following theorem is proved:

$$\int P_l(\mu)d\Omega \int N(z,\mu')F(\mu_0)d\Omega' = F_l N_l(z) \quad (15)$$

We obtain our result by applying this theorem for the case $l = 0$, remembering that, by Eq. 3

$$F_0 = \int F(\mu_0) d\Omega = 1 \qquad (3')$$

We obtain a second equation by multiplying Eq. 13 by μ and again integrating over $d\Omega$, which leads to:

$$\frac{1}{3}\frac{dN_0}{dz} = -N_1 + (1-f)F_1 N_1 \qquad (16)$$

The second term on the right-hand side of Eq. 16 follows immediately from our general theorem (Eq. 15). The terms on the left-hand side may be derived by noting that:

$$\mu^2 = \tfrac{1}{3} + \tfrac{2}{3}P_2(\mu) \qquad (17)$$

This leads to:

$$\int \mu^2 N(\mu,z) d\Omega = \tfrac{1}{3}N_0(z) + \tfrac{2}{3}N_2(z) \sim \tfrac{1}{3}N_0(z) \qquad (18)$$

inasmuch as we have agreed to neglect $N_2(z)$ and all higher-order terms.

We are thus led to the simultaneous total differential Eqs. 14 and 16. These may be reduced to a single differential equation of the second order:

$$-\frac{1}{3(1-F_1)}\frac{d^2N_0}{dz^2} = -fN_0 + S_0(z) \qquad (19)$$

where we have neglected the product fN_1 because both quantities are assumed to be small. Eq. 19 is known as the diffusion equation, and may be written in the more customary form:

$$\nabla^2 N_0 - \frac{1}{L^2}N_0 + \frac{1}{D}S_0 = 0 \qquad (20)$$

Here the quantity L is known as the diffusion length, and is a measure of the mean distance traveled by a particle before absorption. D is the diffusion coefficient and has the value

$$D = \frac{1}{3}\frac{\lambda_s}{1-F_1}V = \frac{1}{3}\lambda_{tr}V \qquad (21)$$

Here the quantity λ_{tr} is called the transport mean free path, and is related to the scattering mean free path by:

$$\lambda_{tr} = \frac{\lambda_s}{1-(\cos\theta)_{av}}, \qquad (22)$$

$$(\cos\theta)_{av} = \int F(\mu_0)\mu_0 d\Omega = F_1$$

Also we have:

$$L^2 = \frac{1}{3\sigma_s^2 f(1-F_1)} = \frac{1}{3}\lambda_c\lambda_{tr} \qquad (23)$$

or $L^2 = D\tau_c$ with τ_c = mean life = λ_c/V, where $\lambda_c = \sigma_c^{-1}$ is the capture mean free path.

Relation Between Point and Plane Sources (*1,2*)

There is actually no loss in generality in solving form 1 of Boltzmann's equation (Eq. 1) one-dimensionally, that is, for a plane source. This follows from the fact that the solution for a point source is expressible as a certain derivative of the solution for a plane source.

Let $g(r)$ be the neutron density $N_0(r)$ which is a solution of Boltzmann's equation for a point source at $r = 0$. Then for any distribution of isotropic sources $S_0(\mathbf{r})$, the corresponding density is given by the integral:

$$N_0(\mathbf{r}) = \int d\mathbf{r}' S_0(\mathbf{r}') g(|\mathbf{r}' - \mathbf{r}|) \qquad (24)$$

In particular, let $S_0(\mathbf{r}) = \delta(z)$, where $\delta(z)$ is the so-called Dirac δ-function defined to be zero everywhere except at $z = 0$, where it is infinite in such a way that:

$$\int_{-\epsilon}^{+\epsilon} \delta(z)dz = 1;$$

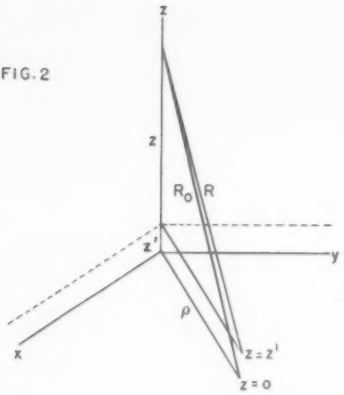
$$\int_{-\epsilon}^{+\epsilon} f(z)\delta(z)dz = f(0) \qquad (25)$$

We see from Fig. 2 that:

$$N_0(z) = \int_0^{2\pi} d\phi \int_0^{\infty} \rho d\rho \int_{-\infty}^{+\infty} dz'\delta(z')g\left[\sqrt{(z-z')^2+\rho^2}\right]$$

$$= \int_0^{2\pi} d\phi \int_0^{\infty} \rho d\rho g\left[\sqrt{\rho^2+z^2}\right] = 2\pi \int_{|z|}^{\infty} R_0 dR_0 g(R_0) = \chi(z) \qquad (26)$$

FIG. 2



with $R_0 = \sqrt{z^2 + \rho^2}$. From Eq. 26 it follows immediately that:

$$g(R_0) = -\frac{1}{2\pi R_0}\frac{d\chi(z)}{dz}\bigg|_{z=R_0} \quad (27)$$

which constitutes the required proof.

Solutions of Diffusion Equation (2)

Plane source in an infinite medium: The general solution is given by

$$N_0(z) = Ae^{-z/L} + Be^{z/L} \quad (28)$$

where A and B are arbitrary constants to be determined by the boundary conditions that the neutron density shall vanish at $z = +\infty$, and that the current shall have the proper discontinuity at the source, with the density continuous through the plane of the source. The regions to the left and right of the source must be treated separately. From the conditions at infinity we have:

$$N_0^{(R)}(z) = Ae^{-z/L}; \qquad N_0^{(L)}(z) = Be^{z/L} \quad (29)$$

where R and L refer to conditions to the right and left of the source, respectively. The continuity of N requires immediately that $A = B$. To determine the magnitude of A, we integrate the differential equation from a point just to the left of the boundary to a point just to the right. Thus we have:

$$\lim_{\epsilon\to 0}\int_{-\epsilon}^{+\epsilon}\left\{\frac{d^2N_0}{dz^2} = \frac{1}{L^2}N_0 + \frac{1}{D}\delta(z)\right\}dz$$
$$= \left[\frac{dN_0^{(R)}}{dz} - \frac{dN_0^{(L)}}{dz}\right]_{\epsilon\to 0} + \frac{1}{D}$$
$$= 0 \quad (30)$$

from which we find the correct solution:

$$A = \frac{L}{2D}; \qquad N_0(z) = \frac{L}{2D}e^{-|z|/L}$$
$$= \frac{3L}{2}(1 - F_1)e^{-|z|/L} \quad (31)$$

The insertion of the absolute value sign in the exponential makes the solution appropriate to both sides of the boundary.

Plane source at $z = z'$ located in a semi-infinite medium, with a boundary to vacuum ($N_0 = 0$) located at $z = 0$: This problem may be solved by the method of images. We add to the solution for a plane source at $z = z'$, the solution due to a sink (*i.e.*, a negative source of neutrons) of equal strength at $z = -z'$. The boundary now lies midway between the two sources, and N_0 automatically vanishes at the boundary because of symmetry. On the basis of such reasoning, we obtain the solution:

$$N_0(z) = \frac{L}{2D}\left\{e^{-|z-z'|/L} - e^{-|z+z'|/L}\right\} \quad (32)$$

where the second term in brackets is due to the image.

Rigorous Solution of Boltzmann's Equation (1)

For simplicity we restrict ourselves to isotropic scattering, that is, to $F(\mu_0) = 1$, and as usual treat the case of a plane source in an infinite medium. With these simplifications, form 2 of Boltzmann's (Eq. 13) reduces to

$$\mu\frac{\partial N(z,\mu)}{\partial z} = -N(z,\mu) + (1 - f)\frac{N_0(z)}{4\pi} + \frac{\delta(z)}{4\pi} \quad (33)$$

We take the Fourier transform of this equation, obtaining:

$$-iy\mu M(y,\mu) + M(y,\mu) = \frac{1-f}{4\pi} M_0(y) + \frac{1}{4\pi} \qquad (34)$$

where $M(y,\mu) = \int_{-\infty}^{+\infty} N(z,\mu)\, e^{+iyz} dz$.

Solving for M and integrating over $d\Omega$:

$$M_0(y) = \frac{(1-f)M_0(y)}{4\pi} \int \frac{d\Omega}{1-iy\mu} + \frac{1}{4\pi} \int \frac{d\Omega}{1-iy\mu} \qquad (35)$$

Evaluation of the integral yields:

$$\int \frac{d\Omega}{1-iy\mu} = \frac{2\pi}{-iy} \ln \frac{1-iy}{1+iy} = 4\pi \frac{\tan^{-1} y}{y} \qquad (36)$$

whence we obtain:

$$M_0(y) = \frac{\dfrac{\tan^{-1} y}{y}}{1-(1-f)\dfrac{\tan^{-1} y}{y}} \qquad (37)$$

Or, performing the Fourier inversion,

$$N_0(z) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} M_0(y) e^{-iyz} dy \qquad (38)$$

This integral may be most easily evaluated by complex integration methods, the details of which are discussed in Appendix B. The result is Eq. 39 (see bottom of page), where ν is a root of the transcendental equation

$$\frac{\tanh^{-1} \nu}{\nu} = \frac{1}{1-f} \qquad (40)$$

The integrated term in Eq. 39 falls off at least as fast as e^{-z}, and actually somewhat faster. This follows from the fact that the lower limit of integration is unity and, at this limit, furthermore, the denominator is infinite. The first term, on the other hand, falls off less rapidly than e^{-z}, since it follows from Eq. 40 that even in the case of very high capture, that is, $f \sim 1$, the upper limit of ν is still 1. Thus there always exists a distance from the source, large compared with a mean free path, at which the second term can be neglected. Actually, if the capture is small, the first term is a good approximation even at distances comparable to a mean free path. In the limit of small capture the secular Eq. 40 may be solved by expanding the inverse hyperbolic tangent in powers of ν. In this way we obtain:

$$\frac{\tanh^{-1} \nu}{\nu} \sim 1 + \frac{\nu^2}{3} \sim 1 + f$$

or

$$\nu = \sqrt{3f} = 1/L \qquad (41)$$

For the approximate solution we have:

$$N_0(z) = \frac{\sqrt{3f}\,(1-3f)}{(1-f)(2f)} e^{-\sqrt{3f}\,z} \sim \frac{3}{2} \frac{1}{\sqrt{3f}} e^{-\sqrt{3f}\,z} = \frac{3}{2} L e^{-z/L} \qquad (42)$$

which, upon comparison with Eq. 31 and Eq. 23, is seen to be identical with the diffusion result when we set $F_1 = 0$ for isotropic scattering and remember that the unit of length is the total mean free path for scattering and capture.

The importance of the exact solution lies in the fact that it suggests a procedure for correcting diffusion theory. In fact, we have seen that far from the source the integral term may be neglected and the first term has the same form as the diffusion solution provided we take for the diffusion length the reciprocal of the correct solution of the secular Eq. 40 and modify the source term so as to give the same coefficient as in Eq. 39.

$$N_0(z) = \frac{\nu(1-\nu^2)}{(1-f)(\nu^2-f)} e^{-\nu z} + \frac{1}{2} \int_1^{\infty} \frac{e^{-\eta z} \dfrac{d\eta}{\eta}}{\left\{\left(1 - \dfrac{1-f}{\eta} \tanh^{-1} \dfrac{1}{\eta}\right)^2 + \left(\dfrac{\pi(1-f)}{2\eta}\right)^2\right\}} \qquad (39)$$

In view of Eq. 27, the solution for a point source in an isotropically scattering medium is, for large r

$$N_0(r) = \frac{\sigma}{V} \frac{\nu^2(1-\nu^2)}{(1-f)(\nu^2-f)} \frac{e^{-r/L}}{2\pi r} \quad (43)$$

$$L = \frac{1}{\nu\sigma}$$

(We have introduced explicitly the dependence on the total cross section, σ, and the velocity V.) By superposing solutions of Eq. 43, we may obtain the asymptotic neutron density due to a general distribution of sources. Thus, if we introduce the function η_0 which satisfies the differential equation

$$-L^2\sigma_c V\nabla^2\eta_0 + v\sigma_c\eta_0 = \frac{1-\nu^2}{1-f}\frac{2f}{\nu^2-f}S(r) \quad (44)$$

then the actual neutron density, N_0, will be asymptotically equal to η_0 far from sources. This is true because the solution of Eq. 44, when $S(r)$ is a δ-function, is of the same form as the asymptotic solution (Eq. 43) of the Boltzmann equation for a point source. Equation 44 embodies a simple recipe for correcting diffusion theory when the ratio σ_c/σ is not infinitesimally small. The recipe is to replace the source S by an effective source S' where

$$S' = \frac{1-\nu^2}{1-f}\frac{2f}{\nu^2-f}S \quad (45)$$

and to use for the neutron current J, the expression

$$J = -L^2\sigma_c V\nabla\eta_0 \quad (46)$$

In terms of J and S', Eq. 44 is

$$\nabla \cdot J + \sigma_c V\eta_0 = S' \quad (47)$$

When σ_c/σ is small, S' is approximately equal to S, and J is given by the simple expression

$$J = -\frac{\sigma_{tr}V}{3}\nabla\eta_0 \quad (48)$$

Equations 46 and 47 represent the actual physical current only in the asymptotic region.

The above arguments strictly justify Eq. 44 only for an infinite medium. The same equation, however, also applies to a finite medium at points not too close to boundaries. In the general case, the solution for the neutron density is made up not only of terms of the form of Eq. 43, but also of homogeneous terms which are solutions of Eq. 44 with $S = 0$. Such terms can be shown to satisfy the rigorous Boltzmann equation in a source-free region far from boundaries. Thus, the recipe given above still applies provided that appropriate boundary conditions are used with Eq. 44. When the boundaries are sufficiently far from the sources, the boundary problem can be considered by itself, and as we shall see later, the boundary conditions can then be formulated in a simple manner. Thus, Eq. 44 provides, in essence, a means of separating the problems of sources and boundaries.

As we have previously emphasized, the solution of Eq. 44 does not accurately represent the neutron density at points close to sources. The nature of the deviations can be seen by studying the rigorous solution (Eq. 39). For sufficiently small z, less than a mean free path, there comes a point where the integral term of Eq. 39 dominates. In fact, the integral behaves roughly as

$$Ei(z) = \int_z^\infty \frac{e^{-t}}{t}\,dt$$

which diverges logarithmically near $z = 0$. This is in sharp contradiction to the diffusion solution where the neutron density remains finite. The infinite density in the true solution originates from particles emitted directly from the source before they have suffered any collisions, the chief contribution coming from particles emitted nearly parallel to the plane. The infinite density is consistent with a finite particle flux because the distribution of velocity becomes increasingly anisotropic parallel to the plane.

In virtue of Eq. 27, the rigorous solution for a point source takes the form:

$$\frac{\nu^2(1-\nu^2)}{(1-f)(\nu^2-f)}\frac{e^{-\nu r}}{2\pi r}+\frac{1}{4\pi r}\int_1^\infty \frac{e^{-\nu r}d\nu}{\left\{\left(1-\frac{1-f}{\eta}\tanh^{-1}\frac{1}{\eta}\right)^2+\left(\frac{\pi(1-f)}{2\eta}\right)^2\right\}} \qquad (45')$$

At very small distances the integral term behaves as $e^{-r}/(4\pi r^2)$ which is just the probability distribution for particles which have made no collisions since leaving the source. This fact suggests a further modification of diffusion theory, namely, replacing the δ-function source by a so-called first-collision source of the form e^{-r}/r^2. An illustration of this procedure is given below.

Corrections to Diffusion Theory Near Boundaries

So far, we have considered the corrections which must be made to diffusion theory to take into account strong absorption and proximity to sources. We now consider the corrections which must be made close to boundaries.

Let us consider the medium which is bounded by the plane $z = 0$ so that there is a vacuum for $z < 0$, and in which the neutrons are diffusing. We shall assume that there is no capture and shall use units in which σ and V are both equal to unity. For large positive z, we can apply diffusion theory, which gives in the present instance:

$$\frac{d^2N_0}{dz^2} = 0 \qquad (49)$$

so that

$$N_0 = az + b = a(z + z_0) \qquad (50)$$

(we assume no dependence on x and y). The current is

$$J = -\frac{1}{3}\frac{dN_0}{dz} = -\frac{a}{3} \qquad (51)$$

We wish to determine the quantity b. Close to the boundary it is important to consider the neutron density as a function of μ. Let us first use approximate arguments of the sort previously discussed. We then write Eq. 11:

$$N(z,\mu) = \frac{1}{4\pi}[N_0(z) + 3\mu N_1(z)] \qquad (52)$$

Since $N_1(z)$ is the current, we must have

$$N_1(z) = -\frac{a}{3} \qquad (53)$$

At $z = 0$ we must apply the boundary condition:

$$N(0,\mu) = 0 \qquad \mu > 0 \qquad (54)$$

This relation expresses the fact that no neutrons return from the vacuum. Equation 54 cannot be satisfied exactly since only the first two terms of the expansion of $N(z,\mu)$ in spherical harmonics are retained in Eq. 52. We, therefore, simply require that the contribution to the current coming from values of μ greater than zero vanishes, *i.e.*,

$$2\pi\int_0^1 \mu N(z,\mu)d\mu = 0 \qquad (55)$$

This yields immediately:

$$\frac{N_0(0)}{a} = z_0 = \frac{2}{3}$$

If, instead of requiring the integral in Eq. 55 to vanish, we had required the integral of some other function of μ times $N(z,\mu)$ from zero to one to vanish, we would have obtained a different value for z_0. Hence, the value of z_0 obtained by the above crude argument is somewhat arbitrary. Fortunately, the problem of the neutron distribution near a plane boundary can be solved rigorously. The procedure is to reduce the problem to an integral equation which can then be solved by Wiener-Hopf methods (*6*, *7*, *8*).

As will be shown in Part II, the neutron density N_0 in a non-absorbing medium which scatters isotropically satisfies the following integral equation:

$$N_0(\mathbf{r}) = \int\int\int (1-f)N_0(\mathbf{r}')d\mathbf{r}'\frac{e^{-|\mathbf{r}-\mathbf{r}'|}}{4\pi|\mathbf{r}-\mathbf{r}'|^2} \qquad (56)$$

(we are assuming that the mean free path is equal to unity). In a plane problem where N_0 depends only on z, Eq. 56 becomes:

$$N_0(z) = \tfrac{1}{2}(1-f)\int_0^\infty E(|z-z'|)N_0(z')dz'$$

where

$$E(x) = -E_1(-x) = \int_x^\infty \frac{e^{-u}}{u}\,du \quad (57)$$

The limits of integration in Eq. 57 are appropriate for the case of a semi-infinite medium which extends from $z = 0$ to $z = \infty$. By Wiener-Hopf methods it can be shown that the solution of Eq. 57 is asymptotically of the form of Eq. 50 with $z_0 = 0.7104$ when $f = 0$. For small z the actual value of N_0 falls below the value given by the asymptotic solution as shown in Fig. 4 (*8*).

At $z = 0$, $N_0 = a/\sqrt{3} = 0.577a$, whereas the asymptotic solution would give $0.7104a$.

The function $N_1(0,\mu)$ is especially interesting since it represents the angular distribution of neutrons emerging from the surface of a scattering medium.

The number of neutrons emerging from the plane per unit time with cos θ between μ and $\mu + d\mu (\mu < 0)$ is:

$$(\cos\theta)N(0,\mu)d\Omega = 2\pi|\mu|N(0,\mu)d\mu =$$

$$2\pi(1-f)d\mu\int_0^\infty e^{-x/|\mu|}\frac{N_0(x)}{4\pi}\,dx \quad (58)$$

The factor $e^{-x/|\mu|}$ under the integral gives the probability that a neutron, after being scattered into the solid single $d\Omega$ at a distance x from the surface, will reach the surface without being scattered again. The integral in Eq. 58 has been evaluated numerically for $f = 1$ using the rigorous solution for $N_0(x)$. An approximate expression, due to Fermi (*9*), can be obtained by assuming that $N_0(x) = a(1/\sqrt{3} + x)$. (As can be seen from Fig. 4, this will make $N(0,\mu)$ too small, especially for $\mu \sim -1$.) We then find from Eq. 58 that

$$N(0,\mu) = \frac{a}{4\pi\sqrt{3}}(1 + \sqrt{3}\,|\mu|) \qquad \mu \leq 0 \quad (59)$$

As it stands, Eq. 59 does not give correctly the neutron density or current at the surface. These errors can be rectified by changing the normalization, but not the form of Eq. 59. If this is done, we obtain

$$N(0,\mu) = \frac{a}{2\pi\sqrt{3}}\frac{1+\sqrt{3}\,|\mu|}{\left(1+\frac{\sqrt{3}}{2}\right)} \qquad \mu < 0 \quad (60)$$

Using this expression, we find

$$2\pi\int_{-1}^{0} N(0,\mu)d\mu = \frac{a}{\sqrt{3}} = N_0(0) \quad (61)$$

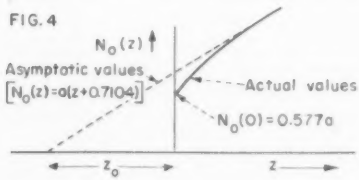
$$2\pi\int_{-1}^{0}\mu N(0,\mu)d\mu = -\frac{a}{3} = J \quad (62)$$

Comparison with the exact solution (*10*) shows that Eq. 60 is correct to 1% for $\mu > 0.1$. For $\mu = 0$, the value given by Eq. 59 is correct, so that Eq. 60 is too high by the factor

$$\frac{2}{1+\frac{\sqrt{3}}{2}} = 1.072.$$

By way of comparison, we note that for photons emerging from a hole in a black body, $N(0,\mu)$ is independent of μ, whereas for neutrons emitted from an isotropic infinite plane source in a vacuum, $N(0,\mu)$ is proportional to $1/\mu$. In the distribution given by Eq. 60, there is a small preponderance of neutrons moving in the forward direction (away from the plane) as compared to the distribution of photons from a hole in a black body.

FIG. 4



The quantity z_0 appearing in Eq. 50 is known as the extrapolated endpoint. When diffusion theory (*e.g.*, Eq. 55) is applied to problems involving a free surface, the boundary condition to be imposed on N_0 is that it vanish, not at the surface, but at a distance z_0 beyond the surface. When this boundary condition is applied in conjunction with diffusion theory, quite accurate results are obtained in many problems of practical importance.

When absorption is present the value of the extrapolated endpoint depends somewhat on f. It turns out that z_0 is roughly proportional to $1/(1-f)$ as is shown in the following table (*11*):

f	0	0.5	0.9	1
$z_0(1-f)$	0.7104	0.7204	0.85	1.000

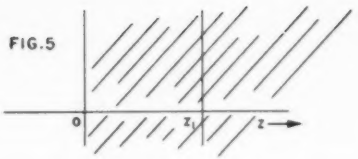
Hence, for reasonably small values of f, $z_0 = 0.71/(1-f)$.

If the units of length are not chosen so that σ is unity, then the extrapolated endpoint is approximately

$$\frac{z_0}{\sigma} \cong \frac{0.71}{\sigma\left(1 - \frac{\sigma_c}{\sigma}\right)} \cong \frac{0.71}{\sigma_s} \qquad (63)$$

From Eq. 63 it may be seen that the extrapolated endpoint is determined primarily by the scattering cross section rather than by the total cross section (remember that cross sections are here measured in cm^{-1}).

We can illustrate the use of the extrapolated endpoint by employing it in conjunction with Eq. 55 to solve the problem of a plane source in a semi-infinite medium. (See Fig. 5.) Assume that the position of the source, z_1, is not too close to the boundary, so that there is a region between the source and boundary which may be considered to be far from both source and boundary. If the source emits one neutron per unit time and area, then the general solution of Eq. 44 is



$$N_0(z) = \frac{1}{2}\frac{1-\nu^2}{1-f}\frac{2f}{\nu^2-f}\frac{1}{\sigma_c VL}e^{-\frac{|z-z_1|}{L}} + Ae^{-\frac{z}{L}} \qquad (64)$$

where A is an arbitrary constant. The constant A must be chosen so that N_0 vanishes at the extrapolated endpoint which is located at the point $z = -0.71/\sigma_s$. This condition gives immediately

$$N_0(z) = \frac{1}{2}\frac{(1-\nu^2)}{1-f}\frac{2f}{(\nu^2-f)}\frac{1}{\sigma_s VL} \times \left\{ e^{-\frac{|z-z_1|}{L}} - e^{-\left(z+z_1+2\times\frac{0.71}{\sigma_s}\right)\frac{1}{L}} \right\} \qquad (65)$$

It should be remembered that this solution (like all solutions obtained by the extrapolated endpoint method) is not valid close to the boundary. In particular, it should not be construed to give any information whatsoever about what happens to the neutron density for $z < 0$.

As a second illustration, we shall compute the neutron density in a nonabsorbing, isotropically scattering sphere with a unit source at the center. Equation 44, together with the boundary condition that the density goes to zero at $R + 0.71$, gives (assuming that the mean free path is unity, and neglecting the correction in the extrapolated endpoint because of the curvature of the surface):

$$N_0 = \frac{1}{4\pi}\left(\frac{1}{r} - \frac{1}{R+0.71}\right) \qquad (66)$$

The result (Eq. 66) can be slightly improved by using a first collision source. The density of neutrons which have not experienced a collision is:

$$N_0' = \frac{e^{-r}}{4\pi r^2} \qquad (67)$$

The density of neutrons N_0'' which have experienced one or more collisions

satisfies the equation

$$-\frac{1}{3}\nabla^2 N_0'' = \frac{e^{-\tau}}{4\pi r^2} \qquad (68)$$

Thus, N_0'' is given by:

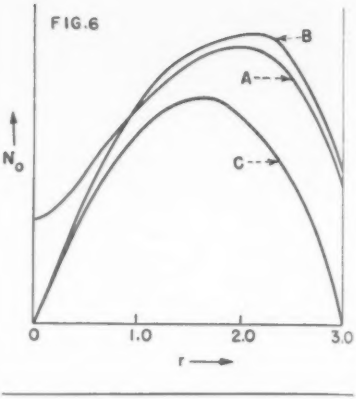
$$N_0'' = \frac{3}{4\pi}\left(\frac{1}{r} - \frac{e^{-\tau}}{r} + \int_r^\infty \frac{e^{-\tau'}}{r'}\,dr'\right) + C \qquad (69)$$

where C is a constant to be determined. The neutron density is the sum of N_0' and N_0''. Since the neutron density must vanish at the extrapolated endpoint $R + 0.71$, we have:

$$N_0 = \left[-\frac{1}{4\pi}\frac{e^{-\tau}}{r^2} - \frac{3}{4\pi}\left(\frac{1}{r} - \frac{e^{-\tau}}{r} + \int_r^\infty \frac{e^{-\tau'}}{r'^2}\,dr'\right)\right]_r^{R+0.71} \qquad (70)$$

In Fig. 6 we have plotted $r^2 N_0$ for $R = 3$ using Eqs. 70, 66, and 66 with $1/(R + 0.71)$ replaced by $1/R$ (curves A, B, and C, respectively). These curves illustrate the errors inherent in elementary diffusion theory near sources and boundaries. In curves A and B the value of N_0 at the surface of the sphere has been reduced to 0.577/0.7104 of the values given by the corresponding formulas in accordance with Fig. 4.

FIG. 6



APPENDIX A

To prove Eq. 15, write both N and F in their expanded forms:

$$\int P_l(\mu)d\Omega \int \sum_{l_1=0}^{\infty}\sum_{l_2=0}^{\infty} \frac{2l_1+1}{4\pi} N_{l_1}(z)P_{l_1}(\mu') \frac{2l_2+1}{4\pi} F_{l_2}P_{l_2}(\mu_0)d\Omega' \qquad (A1)$$

From the well-known addition theorem of spherical harmonics, we know that:

$$P_{l_2}(\mu_0) = P_{l_2}(\mu)P_{l_2}(\mu') + 2\sum_{m=1}^{l_2}(-1)^m \frac{(l_2-m)!}{(l_2+m)!} P_{l_2}^m(\mu)P_{l_2}^m(\mu')\cos m(\phi - \phi') \qquad (A2)$$

Integration over the solid angle $d\Omega'$ involves integration over the angle ϕ', and, since ϕ' occurs in the integrand only through the terms in the expansion A2 for which $m \neq 0$, these terms all integrate to zero, and we are left with the following expression for the integral A1.

$$\sum_{l_1=0}^{\infty}\sum_{l_2=0}^{\infty} \int P_l(\mu)d\Omega \int \frac{2l_1+1}{2} N_{l_1}(z)P_{l_1}(\mu') \frac{2l_2+1}{4\pi} F_{l_2}P_{l_2}(\mu)P_{l_2}(\mu')d\mu'$$

$$= \sum_{l_1=0}^{\infty} \int P_l(\mu)d\Omega N_{l_2}(z)F_{l_2} \frac{2l_2+1}{4\pi} P_{l_2}(\mu) = N_l(z)F_l \qquad (A3)$$

These results are obtained by applying the orthogonality relation:

$$\int_{-1}^{+1} P_k(\mu)P_l(\mu)d\mu = \frac{2}{2l+1}\delta_{kl} \qquad (A4)$$

where the symbol δ_{kl} is the so-called Kronecker δ, which vanishes when $k \neq l$ and is unity otherwise. Equation A3 leads immediately to Eq. 15.

APPENDIX B

It is required to evaluate the integral

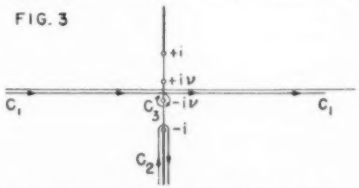
$$N_0(z) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \frac{\dfrac{\tan^{-1} y}{y}}{1 - (1-f)\dfrac{\tan^{-1} y}{y}} e^{-iyz} dy \tag{B1}$$

The integrand is multiple-valued with branch points at

$$y = +i, \qquad y = -i, \qquad y = \pm\infty \tag{B2}$$

Furthermore, the integrand has two simple poles at $y = \pm i\nu$, where ν is the solution of the secular Eq. 40. The situation is illustrated in Fig. 3, where we have taken the branch cuts from $+i$ to $+i\infty$ and from $-i$ to $-i\infty$. The value of $\tan^{-1} y$ increases by π each time a branch point is circumnavigated clockwise. For positive values of z, the contour C_1 may be distorted into the lower half plane in such a way that the integrand remains finite. In this way the contour C_2 is distorted into the two contours C_2 and C_3 as indicated in Fig. 3.

FIG. 3



The integral over C_3 gives

$$-i \text{ Res.} \frac{\dfrac{\tan^{-1} y}{y}}{1 - (1-f)\dfrac{\tan^{-1} y}{y}} e^{-iyz} \qquad \text{at} \qquad y = i\nu \tag{B3}$$

We get:

$$\frac{-i\dfrac{\tanh^{-1}\nu}{\nu}}{\dfrac{\partial}{\partial y}\left[1 - (1-f)\dfrac{\tan^{-1} y}{y}\right]_{y=-i\nu}} e^{-\nu z} = \frac{\dfrac{\tanh^{-1}\nu}{\nu} e^{-\nu z}}{(1-f)\left[\dfrac{1}{\nu(1-\nu^2)} - \dfrac{\tanh^{-1}\nu}{\nu^2}\right]} \tag{B4}$$

Making use of Eq. 40, Eq. B4 becomes:

$$\frac{\nu(1-\nu^2)}{(1-f)(\nu^2-f)} e^{-\nu z} \tag{B5}$$

which is seen to be identical with the first term of Eq. 39.

To evaluate the integral over C_2 we change the variable to $y = -i\eta$. Also we observe that the value of $\tan^{-1} y$ is greater by π on the right-hand side of the branch line than on the left. Furthermore, we note that:

$$\begin{aligned} \tan^{-1} y &= \frac{\pi}{2} - \tan^{-1}\frac{1}{y} \\ -i\tanh^{-1}\eta &= \frac{\pi}{2} - i\tanh^{-1}\frac{1}{\eta} \end{aligned} \tag{B6}$$

Thus we may take

$$\tan^{-1} y = -i\tanh^{-1}\frac{1}{\eta} + \frac{\pi}{2}$$

on the right-hand contour C_2 and

$$\tan^{-1}\eta = -i\tanh^{-1}\frac{1}{\eta} - \frac{\pi}{2} \tag{B7}$$

on the left-hand contour, using the principal value of $\tan^{-1}\frac{1}{\eta}$ in each case. In this way we obtain for the complete contour C_3:

$$\frac{1}{2\pi i}\int_1^\infty \frac{\frac{1}{\eta}\tanh^{-1}\frac{1}{\eta} - \frac{\pi}{2i\eta}}{1-(1-f)\left[\frac{1}{\eta}\tanh^{-1}\frac{1}{\eta} - \frac{\pi}{2i\eta}\right]} e^{-\eta z}d\eta$$

$$-\frac{1}{2\pi i}\int_1^\infty \frac{\frac{1}{\eta}\tanh^{-1}\frac{1}{\eta} + \frac{\pi}{2i\eta}}{1-(1-f)\left[\frac{1}{\eta}\tanh^{-1}\frac{1}{\eta} + \frac{\pi}{2i\eta}\right]} e^{-\eta z}d\eta \tag{B8}$$

Combining the integrands and reducing to a common denominator, we find by straightforward algebra:

$$\frac{1}{2}\int_1^\infty \frac{e^{-\eta z}\frac{d\eta}{\eta}}{\left\{\left[1-(1-f)\frac{1}{\eta}\tanh^{-1}\frac{1}{\eta}\right]^2 + \left[\frac{\pi(1-f)}{2\eta}\right]^2\right\}}$$

which is identical with the second term of Eq. 39 and thus proves the required evaluation.

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URANIUM RESOURCES*

How does uranium occur in nature?

Is there enough uranium to sustain a continuing atomic energy program directed toward both peaceful and military applications?

What is being done in this country to provide a continuing supply of uranium?

These are the questions that arise in an analysis of the mineral engineering aspects of the atomic energy field, and here are some of the answers.

Uranium in Nature

Genetically, uranium occurrences can be divided into four main types:

(1) igneous rocks
(2) hydrothermal vein deposits
(3) sedimentary rocks
(4) deposits of doubtful and perhaps complex origin.

Certain oxidized deposits may be either considered as a fifth type or as a variant of the basic types mentioned.

Uranium occurs in trace amounts in most igneous rocks but is concentrated notably in granitic igneous rocks. It occurs in granites in concentrations in the order and magnitude of from 0.00003% to 0.0007%. Uranium occurs in granites chiefly in the accessory minerals monazite, xenotime, and possibly zircon (*1*).

Granitic pegmatites (*2*) rich in potash feldspar, however, may contain pitchblende, autunite and other uranium minerals in visible amounts. Some pegmatites, like other uranium ores, have been mined in the past for radium (it may be recalled that it is generally considered that radium is in equilibrium with uranium in the ratio of 3.4×10^{-7} to 1). In general, however, pegmatites are not a large potential source of uranium because the average grade is low, seldom exceeding 0.01% U_3O_8, and the tonnage is generally small. There may ultimately be amounts of uranium recovered as a byproduct of feldspar or mica mining.

A decided preference of uranium for granitic rocks and acid pegmatites, and the grouping of some of the hydrothermal uranium deposits around granitic intrusions clearly indicate that granitic magmas are the great primary source of uranium in the earth's crust.

Mesothermal pitchblende veins (*3*) have yielded most of the uranium and associated radium that have been produced in the world. There are two main types of these:

1. Veins mined principally for lead, zinc, copper, gold and silver, such as the veins of Gilpin and Boulder Counties, Colorado. Pitchblende occurs in these as narrow streaks and patches. Veins of this type have contributed little uranium to world supply.

2. Veins containing cobalt and nickel minerals, in addition to other base metals and minor amounts of gold and silver. It is veins of this character that have produced the famous radium-uranium mines of Great Bear Lake in

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* From an address by J. K. Gustafson, manager of raw materials operations, Atomic Energy Commission, at the Massachusetts Institute of Technology on March 9, 1949.

Canada, the Belgian Congo, the Erzgebirge region of Saxony and Bohemia, and the Cornwall district of England. The three most productive uranium-radium mines of the world are Joachimsthal, Bohemia; Eldorado in Canada, and Shinkolobwe in the Belgian Congo. Some of the principal features of these three deposits are the following:

1. Each occurs as veins in pre-Gambrian sedimentary rocks.

2. The ore in each case is pitchblende associated with cobalt. At Eldorado and Joachimsthal nickel, bismuth, and silver are also present, whereas at Shinkolobwe molybdenum, thorium, tungsten, gold, platinum, and palladium are mentioned, as is lead at Eldorado.

3. In each case the principal nonmetallic gangue minerals are quartz and carbonates. Hematite, chlorite, barite, and fluorite also are prominent at Eldorado.

Some hypothermal quartz veins also contain small shoots of pitchblende but are relatively unimportant.

Marine sediments in many parts of the world contain uranium in low concentrations but in concentrations many times that of other rocks (*4*). Notable in this regard are black shales and phosphorites. A number of these formations contain 0.01% to 0.02% U_3O_8, and at least one, the alum shales of Sweden, contain nodules and lenses of a nearly pure bitumen called "kolm" which, according to published figures, contains 0.5% U_3O_8.

Marine uranium-bearing black shales are characterized by abundant organic matter and sulfides and by small content or absence of carbonate material. The beds richest in uranium occur in thin formations of pre-Mesozoic age. The mineral or compound containing the uranium in these black shales has not yet been identified.

Probably all marine phosphorites contain some uranium. Phosphetic nodules in many marine black shales also contain concentrations of uranium. Although the form in which uranium occurs in phosphorites is unknown, the fact that uranium increases in a general way with increase in phosphate content suggests that the uranium may occur in the space lattice of the phosphate mineral. The phosphorite formations, like the black shales, are characteristically thin and are generally associated with the unconformities or diastems. These facts of occurrence have indicated to the men who have studied them that these uraniferous formations were deposited in large basins adjacent to low, stable, land masses during periods when mechanical erosion was at low ebb and chemical conditions in the sea water inhibited the formation of lime deposits. Very possibly the marine sediments derived from granitic land masses are the most highly uraniferous.

Two main types of uranium deposits are not yet properly classifiable. These are the vanadium-uranium ore deposits occurring on the Colorado Plateau (*5*) and in lesser amounts in other parts of the world, and the Witwatersrand gold-bearing conglomerates of South Africa.

The carnotite and roscoelite-type uranium-vanadium ores of the Colorado Plateau occur as small tabular or lenticular deposits impregnating the flat-lying Morrison sandstone and Entrada sandstone of Jurassic age and the Shinarump conglomerate of Triassic age. They are widely but spottily distributed over an area nearly 200 miles in diameter. The long axis of the deposits, many of which do not contain more than several hundred tons of ore, are nearly parallel to bedding, but the ore does not follow the beds in detail. The mineralogy of the ores is very imperfectly understood. The deposits contain several times as much vanadium as uranium. The bulk of the vanadium occurs in extremely fine-grained minerals of micaceous habit, probably belonging to the hy-

drous mica group of clay minerals. The principal mineral has been recognized tentatively as roscoelite. The principal uranium-bearing minerals are thought to be carnotite and tyuyamunite, sometimes called calcium carnotite. The theories of ore deposition subscribed to by most geologists are either: (*a*) that the ore was precipitated from ground water after the enclosing sands had accumulated, or (*b*) that the vanadium-uranium content was deposited during deposition of the enclosing rocks but was widely distributed in them and has experienced considerable subsequent movement and reconcentration by ground water. Small variations in bedding and sedimentary features of the rock appear to have guided the movement of ore solutions with resulting concentration of the ore. Fossil logs and twigs are often completely replaced by ore and are further evidence that carbonaceous material generally is a precipitant for uranium. These types of ore bodies show a considerable range in both uranium and vanadium content and also a considerable range in the uranium-vanadium ratio. Much of the ore contains anywhere from 0.1 to 0.3% U_3O_8 and from 0.5% to 2.5% V_2O_5, although small tonnages of very much higher grade ore are mined from time to time. These deposits constitute the largest readily available source of uranium in the United States but a source greatly inferior to the high-grade hydrothermal pitchblende deposits of other countries.

Recently along the southwest and western edge of the Colorado Plateau numerous new prospects of uranium (carnotite)-copper ores in the Shinarump conglomerate have been found which are similar in size and occurrence to the uranium-vanadium ores. Whether or not these will be important producers is not yet clear.

The Witwatersrand reefs are extensive beds of metamorphosed quartz conglomerate. It has been known for many years that the gold ores contained small amounts of uraninite, but only recently has the possibility of byproduction of uranium from this source become apparent. A controversy has raged for many years over whether the gold ores had a hydrothermal or a sedimentary origin. The origin of the uranium in this conglomerate is equally in doubt but may not necessarily be the same as that of the gold.

In Canada where glaciation stripped off any old oxidized soil covering that existed, primary uranium minerals, only slightly oxidized, occur at the surface. In countries, however, like Africa and Australia, primary products are oxidized to depths of hundreds of feet. Here pitchblende, which is a black heavy metallic mineral, has been converted in the zone of oxidation to brightly colored secondary minerals, such as torbernite, a bright emerald green hydrated phosphate of copper and uranium; autunite, a lemon yellow hydrated phosphate of calcium and uranium; or carnotite, the canary yellow hydrated vanadate of potassium and uranium. As is the case with copper ores, a flamboyant colorful outcrop does not necessarily mean good primary ore in depth.

Supply of Uranium

The consumption of about one pound per day of U^{235} in fission generates heat at a rate equivalent to approximately 450,000 kilowatts, which is the amount of heat that would be obtained by burning about 1,300 tons of coal per day. Even with low efficiency, U^{235} is obviously a very potent source of power. Although, in naturally occurring uranium only one part in 140 is the fissionable isotope U^{235}, by a process known as "breeding," it is theoretically possible to combine some additional U^{235} with natural uranium and to convert the non-fissionable U^{238} into fissionable plutonium.

By this process, to quote from J. R. Menke's "Nuclear Fission as a Source of Power" (*8*), "for each pound of fissioned (burst) U^{235} together with one pound of ordinary uranium we get (*a*) about ten million kilowatt hours of energy in the form of heat, (*b*) about one pound of fissile element (*e.g.*, plutonium) and (*c*) about one pound of new radioelements." This is about three million times the energy released by the burning of an equal weight of coal. This strange process of eating your cake and having it too is theoretically applicable not only to U^{238} but also to thorium, the use of which would greatly increase available atomic fuel.

The experts, in groping for an estimate of the amount of uranium available, have approached the problem from two directions.

One approach is to take the average uranium content of a great many samples of rock of different kinds and to conclude that the resulting figure of four parts per million or 0.0004% is the amount of total uranium available in the earth's crust. On this basis, the answer is about 1.5×10^{12} tons of uranium in the one mile layer of earth's crust not covered by water. It can be pointed out on this line of reasoning that uranium is 1,000 times as plentiful as gold, 100 times as plentiful as silver, and almost as plentiful as lead or zinc. Such an approach is meaningless except to indicate that uranium is an important material in the earth's crust and, accordingly, that it probably was present in the right places fairly often when geological concentrating processes were at work forming ore bodies. What counts in terms of available uranium is economically exploitable concentrations.

The second approach is to take some prewar figures of uranium production, or more generally of radium production, calculate the amount of associated uranium from these, and assume that these figures and the ore reserve data of that same prewar period are a valid measure of the potential production of uranium in the future. This is perhaps an equally fallacious approach for the following reasons:

1. Even up-to-date ore reserve data, whether for copper, gold, lead, uranium or other metals, mean very little unless it is realized that such estimates merely prove that only a few years of production are blocked ahead. It is not economically justified for most mining companies to spend more money in development than is necessary to maintain ore reserves for more than one to five years. It is often bad business to make capital investments in development work that will not yield returns for a long time to come. Moreover, in this country mining companies have to pay taxes on their proven reserves, and this taxing policy inhibits unnecessary blocking out of reserves. Petroleum figures also have periodically led experts to predict early exhaustion of our oil reserves. Yet, the American Petroleum Institute figures show an annual increase in the known reserves of petroleum in this country almost every year for a decade or more despite increasing rates of production.

2. Also ignored or underrated in this approach is our growing technology and its future application to low-grade uranium sources. Most metals have gone through or are going through a cycle where high-grade deposits are at first the only commercial deposits, and then gradually large low-grade deposits yield to man's technical ingenuity and become important producers. The "mining" of sea water during the war as an "ore" of magnesium is probably the most spectacular example of such a development. The ore reserve-production data approach thus omits from consideration the vast tonnages of marine sediments or by-product production from the South African gold mining industry. That

this latter may be a serious omission is indicated by the statement made in a recent speech by the Minister of Industrial Production of the Union of South Africa, who said, "We believe that we are able to say that the Union of South Africa may produce more uranium than any other country of the world."

3. Neglected by this approach also is the great surge of prospecting for uranium that is going on in all parts of the world. Dr. W. F. James of the Advisory Staff of the Canadian Atomic Energy Commission, for instance, recently announced that already three new properties in Canada are virtually certain to come into production and that there are three or four promising new prospects in addition (*6*).

The commercial aspect of uranium should be kept in mind. The average uranium content of some granites is of the order of magnitude of 0.22 ounces per ton of rock. If we had that much gold in a large granite mass, we would consider it a very profitable mine. Of course, the price of gold is roughly 16 times that of uranium and, generally speaking, it is easier to extract. Nevertheless, if uranium extraction technology improves greatly and if uranium is ever needed badly enough, *i.e.*, its price is high enough, there will be a lot of it available.

It is pertinent to inquire, "What is uranium worth today to a mining company?" There may some day be established a world price for uranium comparable but of different magnitude to the price for gold, but at present there is no such price. Foreign uranium is purchased at negotiated prices. The U. S. Atomic Energy Commission has established for domestic uranium a ten-year minimum price of $3.50 per pound of contained U_3O_8 in a high-grade product, the Canadian Government has established a roughly equivalent minimum price of $2.75 per pound of contained U_3O_8 in Canadian ores and concentrates containing at least 10% U_3O_8 f.o.b. railroad, and the United Kingdom Ministry of Supply has offered to buy all uranium ores and concentrates produced in the Colonial Empire during a ten-year period at a minimum price of approximately $2.78 per pound of contained U_3O_8 delivered f.o.b. ocean port. From this it is readily apparent that even a narrow pitchblende vein means high-grade ore at these prices.

Because uranium is everywhere controlled by the governments of the countries in which it exists, it is unlikely that it will ever appear on free competitive world markets in the sense that lead or copper does. In this connection, the fact is worth noting that the AEC's 10-year guaranteed minimum price for domestic uranium is in fact a minimum price. Where larger quantities are involved than the small lots for which this price was established, or under special circumstances, the Commission is prepared by negotiation to establish higher prices which will take into consideration special milling and refining costs, *etc.*

There is, moreover, at the present time a "subsidy" price in connection with our Colorado Plateau ore-buying program. There is a base price (including development allowance) of $2.50 per pound of contained U_3O_8 in ores containing 0.2% U_3O_8, but payment is also made for the V_2O_5 content at $0.31 per pound, and a haulage allowance of $0.06 per ton mile is allowed up to a limit of 100 miles. Additional premiums are allowed for higher grade material.

The estimate of future uranium supplies may be too pessimistic. For instance, there are those who, basing their predictions on mining experience with other metals, think that there will be new high-grade uranium producers found, that the old producers will last longer than people think, that there will

be significant uranium production from low-grade ores that are not now even considered ore, that the U. S. can get large amounts of thorium when and if needed, and that there will be enough source material to permit the atomic energy business to expand considerably and to go on for generations.

U. S. Uranium Program

During the war, the Manhattan District purchased uranium from Canada and the Belgian Congo and extracted uranium from accumulated tailings of past vanadium operations on the Colorado Plateau. When the AEC took over the atomic energy project, it continued to buy foreign uranium, which to this day constitutes a high percentage of our total plant feed. On April 11, 1948, it announced this three-point program to stimulate the discovery and production of domestic uranium by private competitive enterprise:

1. Government guaranteed 10-year minimum prices of $3.50 per pound of contained U_3O_8 for small lots of domestic refined uranium, and of $3.50 per pound of recoverable U_3O_8 less refining costs for small lots of ore or mechanical concentrates assaying at least 10% U_3O_8, both prices f.o.b. shipping point.

2. A bonus of $10,000 for the discovery of a new deposit and production therefrom of the first 20 tons of uranium ore or mechanically produced concentrates assaying 20% or more U_3O_8.

3. Guaranteed minimum price for uranium-bearing carnotite-type or roscoelite-type ores of the Colorado Plateau area for the period ending June 30, 1954 (Circular No. 3, which extended through April 11, 1951, and Circular No. 4, which extended through June 30, 1949, were combined in new Circular No. 5 on February 1, 1949). Although no important change was made in the pricing provisions of Circulars No. 3 and No. 4, several adjustments were made, and the period of the guarantee was extended for approximately three years in order to attract capital and mining development.

The Commission also has extensive exploration activities which are carried out in large part by the United States Geological Survey (*7*).

An equally important part of the Commission's study of uranium resources is its research program to develop new processes which can treat economically very low-grade uranium materials. Incidental developments of this program already include new and improved analytical techniques and instruments.

The AEC licenses all transfers of source material after its separation from the place of deposit in nature. (The term "source material" means any material except fissionable material containing 0.05% or more of uranium and/or thorium). Consequently, anyone buying or selling uranium or thorium ore after it has been mined must apply to the Commission for a license. Distributors and processors of source material are also required to fill out a simple form each month so that the Commission can keep track of where it is and where it goes.

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CRYSTAL COUNTERS—II

Although most facts about crystal counters can be explained, it is still not known what makes a material a counter. The author analyzes experimental results in this concluding article. Crystal counters will find great use as gamma-ray detectors because of their high stopping power. They have good linearity and resolving times which are faster than those for most counters. However, polarization is a serious handicap. There is a need for, and it is hoped that, a new material will be developed to serve as a room-temperature counter

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THE PUBLISHED MATERIAL in the field of crystal counters, apart from the work of van Heerden, is quite fragmentary at the present time. In considerable degree, the same will necessarily have to be true for this report. The results of the Princeton group may be given here on silver halide counters, but there is no satisfactory comparison which can be made with the work of others. The situation is similar in the case of diamond and other room temperature counters. It is hoped that these gaps in the literature will be closed in the coming years.

It was stated in Part 1 that there are at least two fundamental quantities that are of importance for each type of crystal counter, *viz.*, (*a*) the electron mobility in the crystal and (*b*) the energy per ion pair. Experimental measurements of the former can be compared with theory whereas, at the present time, this is not the case for the latter.

It would be highly desirable, and would serve as a stringent check on theory, if the temperature dependence of the electron mobility were determined experimentally. Such a study might have important consequences for the quantum theory of ionic and non-polar crystals.

Mobility Measurements

The experimental measurement of mobility may be made by observations of the transit time (rise time of the pulse) required for electrons to travel from the negative to the positive electrode. For accurate measurements it is required: (*a*) that secondary electrons start from the negative electrode, (*b*) that most of the electrons travel to the positive electrode without being trapped, and (*c*) that the trap distribution be homogeneous. The first two conditions may be satisfied if alpha or beta particles, both of which have only a short range in the crystal, and a saturation field are used. In this case, if τ is the transit or rise time, w the drift velocity, d the thickness of the crystal, E the field strength, and v the mobility, then

$$\tau = d/w = d/vE \qquad (22)$$

and τ should be observed to vary inversely as E. Therefore, $1/\tau$ should

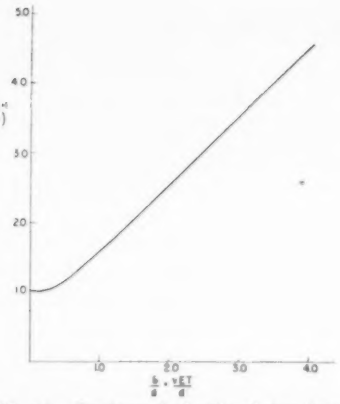

FIG. 14. Reciprocal of ratio of rise time τ °of pulse to free time T before trapping, plotted against δ/d. is proportional to the electric field

vary directly as E. The experimental conditions are generally arranged to meet these conditions and to satisfy relationship (22). However, the assumption implicit in (22) is that the time T during which an electron remains free is large compared with τ. If this is not so, then there is a more accurate, and indeed, more general expression for τ, as shown below.

In general, τ will be the ratio of the average distance moved by the cluster of electrons in a single pulse to the drift velocity of the cluster. Now, the average distance $\bar{x}$ may be taken from Eq. 7 in Part 1 as

$$\bar{x} = dF\left(\frac{\delta}{d}\right) = vET(1 - e^{-\frac{d}{vET}}) \qquad (23)$$

$$\tau = \bar{x}/Ev = T(1 - e^{-\frac{d}{vET}}) \qquad (24)$$

The reciprocal of τ/T is plotted against the variable vET/d in Fig. 14. We see that, if T or E is very large, the limiting value of τ is given by Eq. 22. However, if T or E is very small, then $\tau = T$, which is a result to be expected.*

* This behavior was first pointed out by K. G. McKay (*19*).

In this case a value of the mobility cannot be obtained from the measurements. A satisfactory test of the validity of the mobility measurement may be obtained by checking the data against an equation of type (24). In fact, it is possible, by careful measurement, to make a determination of v in this way. This determination is, of course, equivalent to the determination of δ, if T is known.

It must also be established that d is the proper distance to take for the movement of the electrons. Thus, a barrier which might cause trapping must not be present. This consideration means that the saturation pulse size must be equivalent to the charge carried by the full number of electrons, $n_0 = H\psi/\rho$ (see below).

Energy per Ion Pair Measurements

With a knowledge of the energy H of the particle and the capacitance of the crystal-vacuum tube input circuit, the energy per ion pair may be measured simply by finding the magnitude of the voltage pulse. Thus, from

$$V_{eff} = \frac{n_0 e}{C} F\left(\frac{\delta}{d}\right) \qquad (7a)$$

we obtain, when $F(\delta/d) = 1$, $n_0 = \frac{CV_{eff}}{e}$; by studying the shape of the experimental saturation curve corresponding to Eq. 7 we can also determine n_0 by extrapolation. Thus we determine how many free electrons are produced, and knowing the energy H of the entering particle, we obtain

$$\epsilon = H/n_0 = \rho/\psi = \text{energy per ion pair} \qquad (25)$$

Usually an artificially produced standard pulse is applied to the input circuit and the oscilloscope deflection thereby calibrated. The voltage amplitude of an unknown crystal pulse can be found by comparison with the calibrated pulses. The input capacitance and crystal capacitance are found either together, or separately and then added.

Most troubles arising from unknown trapping of electrons in the crystal tend to give too high a value for the energy per ion pair. The lowest value tends to be the most reliable, other things being equal. It is probably worthwhile, in such experimental studies, to use a collimated beam of particles and to examine different regions of the crystal to see whether the value is constant.

The foregoing discussions are valid when, as in diamond, the holes move or when, as in AgCl at low temperature, the holes are stationary. In the first case, the holes are formed near the negative electrode and therefore contribute a negligible amount to the pulse, if the crystal thickness is large enough. In the second case the holes contribute nothing as they do not move.

For the measurements of energy per ion pair it is certainly best to use monoenergetic particles, such as beta particles from a beta-ray spectrograph (van Heerden). However, if a beta-ray spectrograph is not available, the measurements can be made with a radioisotope, such as P^{32}, whose maximum beta energy is known. One then studies the largest pulses by selection with a discriminator. Absorbing foils can be used to reduce the energy.

Studies of energy per ion pair (and mobility) can also be made by the use of gamma rays as the source of radiation. However, attention must be focussed again on only the largest pulses. For gamma-ray work see reference (*18*). Determinations of mobility made with beta or alpha particles are recommended since simple collimation of the radiation is possible and the conditions of the experiment are under better control.

In the case of energy per ion pair measurements, van Heerden has indicated that alpha particles give pulses much less than their energy would indicate, if the energy per ion pair for a beta particle is used as standard. He suggested that intense local fields might affect the movement of the electrons away from the site of ionization. However, the results of Yamakawa, on alpha particles produced within the body of a mixed crystal by slow neutron capture (LiBr-AgBr), indicate that alpha pulses are larger than gamma pulses of energy about 2.0 Mev. Although the measurements are rough the energy per ion pair does not appear to be significantly different from the value obtained for beta particles. It is therefore possible that surface effects are of importance in alpha particle determinations of energy per ion pair. In at least some diamonds, this does not appear to be so.

Mobility, Energy per Ion Pair: Experimental

Room temperature counters. In the case of diamond, McKay (*19*) has indicated a value of $T = 0.04$ microseconds from results on conductivity induced by electron bombardment. An upper limit for the rise time, this result depends on an assumed value of mobility of 156 cm^2 per volt-sec calculated by Seitz for diamond. Temperature studies of the rise time and energy per ion pair in diamond would be valuable for understanding the counter mechanism, but have not yet been made. Studies of rise time are difficult because of the small values.

McKay gives a value of 10 ev for the energy per ion pair in diamond. Ahearn has given the same value, obtained from his studies with alpha-particle pulses.

J. R. Platt reported at the Rochester conference (*70*) that, for a 0.2 mm gap, the rise time of alpha-particle pulses in CdS was less than or equal to 0.2 microsecond. As studies of rise time versus electric field were presumably not made, it is not possible to draw further conclusions from this result. The energy per ion pair, which can be estimated from the results of Kallmann and

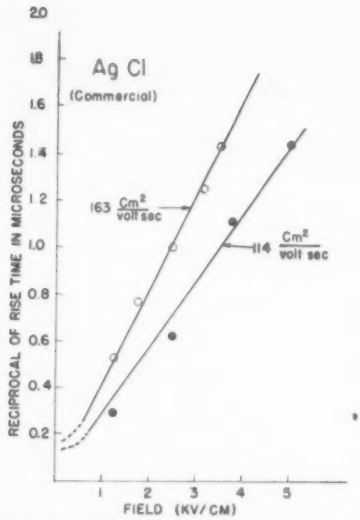

FIG. 15. Experimental data of rise time in AgCl crystal counter plotted against electric field. Curves are similar to theoretically expected prototype of Fig. 14. Data refer to temperature of 77° K, and were taken by Milton and Hofstadter

Warminsky (*48*), is in the neighborhood of 10 ev, as in diamond.

Low temperature counters. (*a*) *Liquid and solid argon*—In liquid argon (*42*) the observed pulses appear to be due to primary effects; the number of electrons produced corresponds to the energy of the ionizing particle divided by about 25 ev. This result has been obtained at a voltage gradient of about 10,000 volts per cm, which is sufficient to give saturation of the pulses. Mobilities of the order of 40 cm² per volt-sec were found by Hutchinson. In solid argon, Hutchinson has found very large pulses—12 times the size of pulses in the liquid—which he attributes to electron multiplication. To obtain the large pulses in the solid, voltage gradients of more than 13,000 volts per cm were used. Polarization occurs in the solid. (See page 34 for further results on solid argon.)

(*b*) *Halides*—The early results (*11, 18*) on AgCl showed that, with commercial samples, the mobilities were in the range of 80 to 400 cm² per volt-sec at 77° K, and the lowest energy per ion pair was about 14 ev. Van Heerden did not measure the mobility but found that the energy per ion pair was 7.6 ev. The discrepancy in values for energy per ion pair can be ascribed to the fact that the commercial material was probably not a single crystal and that barriers and strains were present (see Fig. 8, in Part 1). In fact, it was a discrepancy such as this which partially accounted for the decision to grow synthetic single crystals under known laboratory conditions.

Other data on mobility in AgCl, not published, were obtained by Milton and the writer for a 0.4 cm crystal and are shown in Fig. 15. It is observed that different results were obtained for two different runs, giving mobilities of 163 and 114 cm² per volt-sec. The reasons for the difference in values are unknown although they may be connected with different strains and perhaps slightly different temperatures. The results should be quite sensitive to temperature. An average of the two runs gives 139 cm² per volt-sec.

It is noticed that in both cases the experimental points lie fairly close to straight lines indicating that the assumption $T \gg \tau$ is reasonable. The value 139 cm² per volt-sec lies in the neighborhood of the values reported in (*18*), although it is not as high as the highest value (400 cm²/volt-sec) reported there. The measured values are not in good agreement with the theoretical value, about 1600 cm² per volt-sec, given in Table 1 (p. 9, Part 1).

It must be pointed out that there are alternative values of the characteristic temperature θ which may be substituted in the Fröhlich-Mott expressions. The temperature θ depends on an eigenfrequency of the crystal lattice. In

AgCl there are two restrahlen* from which we might expect two eigenfrequencies. If one uses the highest value ($\sim$ 95 microns) one obtains a mobility of 950 cm² per volt-sec, quite high when compared with experiment.

There is a further uncertainty to be considered. The room temperature values of dielectric constant have been used in evaluating the temperature θ, whereas the low temperature values should be used. However, from the insensitive way in which the dielectric constants enter into θ it is not felt that a very serious change would be produced in the mobility values by using the room temperature constants. The eigenfrequencies of Table 1 have all been taken as equal to the room temperature values.

Commercial AgCl material was used in these experiments and since the purity and condition are unknown it is likely that the observed discrepancy is not serious. However, this point should be cleared up by further studies on pure single crystals. It would be most desirable to measure the mobility at different temperatures, thus providing data for comparison with the theory at more than one point.

The results of Yamakawa for AgBr are in better agreement with theory and offer satisfying justification for growing synthetic crystals. Yamakawa's results which were taken in sequence at progressively larger field strengths, are reproduced in Fig. 16. The sharpening of the pulses is very clear. It will also be noticed that the slope of the rise is reasonably constant, particularly for higher field strengths, indicating a constant drift velocity over the thickness of the crystal. The shape of this rise may give information concerning the homogeneity of the crystal.

Two samples of AgBr which were studied yielded mobility values of 208 and 213 cm² per volt-sec (see Fig. 17). The distance moved by secondary electrons in the crystal was corrected for the slight penetration of the beta particles. The internal agreement is quite satisfactory and is probably a little better than the data would indicate. The theoretical value of v in this case is 296 cm² per volt-sec, thus giving an agreement that is considered reasonable at the present time.

Yamakawa's figure for energy per ion pair in AgBr is 5.8 ev and was obtained by the usual extrapolation to large field strengths. This figure is quite low, even lower than the 7.6 ev

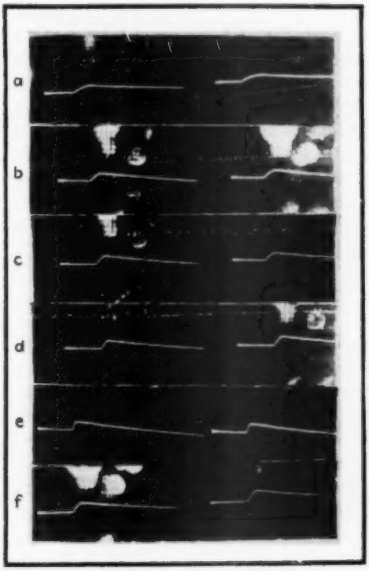

FIG. 16. Shape of typical AgBr crystal counter pulses due to energetic beta particles from P^{32}. Photographs were taken by K. A. Yamakawa. *a*, *b*, *c*, *d* refer to electric fields of 1870, 2730, 3640 and 4550 volts/cm, respectively. Traces *e* are due to an artificial pulser with faster rise time than amplifier-delay line rise time. Traces *f* refer to radium gamma-ray pulses observed at a field strength of 3640 volts/cm

* The restrahlen values are obtained from H. Rubens. *Sitzber. preuss. Akad. Wiss. Physik. math. Klasse*, p. 513 (1913). The eigenfrequencies are slightly different from the restrahlen and are shifted towards longer wavelengths.

value for AgCl. Although this measurement should be repeated in other laboratories, it is not unreasonable that the energy per ion pair is less in AgBr than in AgCl as the optical absorption limits for these two materials are 3.95 and 4.88 ev, respectively (*58*, *59*). It is to be expected that the energy per ion pair will be less if the absorption limit is smaller. The value of ψ for AgBr is therefore about 0.6, as in AgCl.

Polarization: Experimental

Liquid and solid argon. Davidson and Larsh (*41*) have noted that polarization effects should not appear in liquid argon. In solid argon Hutchinson has found polarization phenomena in which pulses continue after removal of the applied field and have the same sign as observed with the field on. As Hutchinson has pointed out, the sign of the pulses may be connected with the secondary ionization effects, although no definite picture has been given.

It has been pointed out that when gamma rays are counted (and the holes are immobile) pulses of both signs may be expected after the applied field has been removed. We do not know whether our simple model applies to solid argon. Among other reasons, our model applies to a single crystal, whereas there is a good possibility that the cooling procedure used in solidifying the argon has resulted in formation of a polycrystal. However, if our ideas are correct, the sign of pulses observed by Hutchinson may be due to the gamma rays converted in the region of the crystal to the left of center in Fig. 5 (Part 1). If electron multiplication does occur it may be expected that positive space charge density will increase to the right (and not remain constant) as the electrons drift to the right and make secondaries in the electrical field. Thus, the point of zero polarization field is shifted to the right. The majority of observed pulses would have the same sign as with field on, which is Hutchinson's result.

Halides. It will be noticed that if: (1) beta particles are used, (2) the crystal is free of barriers, and (3) a homogeneous distribution of traps is assumed, then $a = d - b$ and the expressions

$$0.5\,\frac{n}{1 - a/d}, \qquad 0.8\,\frac{n}{1 - a/d}, \qquad 0.9\,\frac{n}{1 - a/d} \qquad (19a)$$

will depend on the factor nd/b. When b/d is small, as it is in the example on p. 14 (Part 1), the number of events, N_f, required for the field to drop to say $\frac{1}{5}$ of its original value, depends very sensitively on b. Selection of an average value of b does not weigh the effect of an energetic beta particle as much as is really necessary, for an energetic beta particle penetrates farther into the crystal and produces holes deeper within the crystal than a softer beta particle. Therefore, our calculation will err on the side of allowing N_f to be larger than it really is. This effect will not change N_f by more than a factor of about 2. In counting all events equally, this effect is compensated for to some extent by the backscattering of some of the softer beta particles which do not leave all their energy in the crystal and hence do not produce as much polarization.

Yamakawa has studied the effect of polarization produced by beta particles of P^{32} in a sample of AgBr for which the effective area A is 0.4 cm², V_0/d = 3,300 volts/cm, d = 0.6 cm, K = 13 and $\epsilon \cong 6$ volts/ion pair, b = 0.021 cm. By means of a ten channel discriminator (*55*)* Yamakawa also investigated the distribution of pulse sizes as the polarization gradually built up. The

* The ten channel discriminator is an instrument which sorts pulses into ten channels and counts the pulses in each. Therefore differential bias curves are taken directly by it. The channels were ten volts wide in the present use.

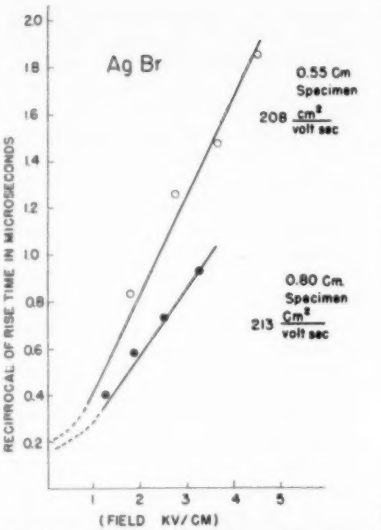

FIG 17. Experimental data on rise time in AgBr crystal counters. Data were taken at 77° K by K. A. Yamakawa

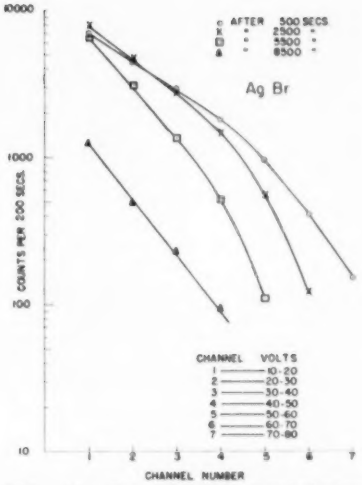

FIG. 18. Ten channel discriminator curves of AgBr pulses due to P^{32} beta particles. The lower curves show the large drop-off in counting rate as the crystal polarizes. Data taken by K. A. Yamakawa

experimental curves are shown in Fig. 18. It can be seen that the number and size of pulses drop drastically after 8,500 seconds of counting. This may be taken to indicate that the electric field has fallen to such a value that saturation no longer exists. The behavior is therefore as described previously where the pulses are now to be considered in the region of the knee of the curve of Fig. 3*a* (p. 7, Part 1).

Counting all the pulses above 10 volts up to time 8,500 secs one obtains 5.6×10^5 pulses. It is difficult to know how many small pulses, below 10 volts, should be added to this result. By extrapolating the measured curves to zero* we can estimate this number to be about 2.7×10^5. When polarization effects are important, the total number of counts is of the order of 8.3×10^5. The theoretical value $N_{0.2}$, computed from the expressions in (19*a*), indicates that about 2×10^6 counts should be observed to obtain a drastic reduction of pulse size. Allowing for the greater importance of large pulses we may take as a rough figure 1×10^6 pulses, which is the correct order of magnitude. Better agreement cannot be sought under the conditions of the experiment, for edge effects of the electric field have not been taken into account, and homogeneous distribution of traps in the crystal and no cracks or other barriers have been assumed. With very rough data and crude approximations, the agreement is quite satisfactory.

Suggested improvements for further work are the use of a large crystal and the employment of a collimated beam of beta particles directed at a small region in the center of the crystal.

* It may be noticed that the curves do not appear to have the familiar Fermi shapes. Edge effects and backscattering in source and detector are probably the principal reasons for the observed shapes.

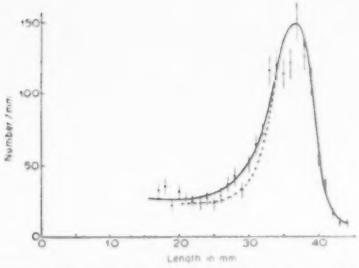

FIG. 19. Pulse distribution, observed by van Heerden, due to homogeneous beta particles of energy 0.4 Mev. The tail at the left is due primarily to back-scattering of beta particles. The dashed curve shows what is expected theoretically

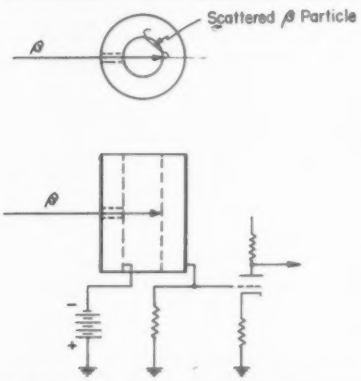

FIG. 20. Suggested shape of crystal counter to avoid the tail of the pulse distribution of Fig. 19

Along theoretical lines the calculation can be improved by use of greater care with the distribution of positive sites produced by the beta particles within the crystal close to the negative electrode, and also by consideration of the negative space charge.

It is clearly possible to perform further interesting experiments on polarization effects in crystals with the use of alpha, beta and gamma rays. As indicated previously, different behavior may be expected for all three types of radiation. The large range of dielectric constants available (silver halides ~ 12, thallium halides ~ 35) makes other checks possible.

Efficiency

Efficiencies are hardly mentioned in the literature. However, Ahearn (*60*) has stated that the efficiency of a diamond counter increases rapidly with increasing voltage gradient and approaches saturation at about 4,000 volts per cm. The highest efficiency observed was 60%. Efficiencies for beta particle counting have not been published for any crystals.

An efficiency figure of 13% has been given (*13*) for a crystal of AgCl 4 mm thick for radium gamma rays. As scattering from chamber walls is probably important and the number of gamma rays from radium uncertain, the quoted results are not to be taken as indicating more than the order of magnitude. Counts of the order of amplifier noise must be discriminated against and therefore the counting rate will, in general, be lower than the expected rate calculated from known cross sections. Efficiency figures are likely to become reliable as soon as plateaus in counting rate are observed, as in Ahearn's results described above.

Linearity

The equation

$$Q_{eff}/n_0 e = F\delta/d \qquad (7)$$

shows that the pulse size is proportional to n_0, which in turn is proportional to the energy of the incident beta or alpha particle. The crystal counter is therefore a linear device, that is, one in which the pulse height is proportional to the energy of the incident particle. Van Heerden showed that this theoretical expectation is verified in practice. Figure 19 is reproduced from his thesis (p. 64). The curve shows that when almost monoenergetic beta particles of energy 0.4 Mev strike the crystal the resulting distribution of pulse heights

has a strong maximum. The width of the maximum is due to beta particle inhomogencity, statistical deviations, amplifier noise, etc. The low energy tail is believed due to backscattering or reflection of the beta particles.

Van Heerden's crystal was a flat plate. Beta particles reflected from the surface may lose only a part of their energy in the crystal plate and so give the smaller pulses found in the observed tail. In principle the tail caused by such reflections could be almost entirely eliminated by making the crystal in the shape of a hollow cylinder with a small entrance hole for the electrons. Figure 20 shows a sketch of this possibility. In this case only those electrons scattered very close to 180° will be lost. All others are recovered. It is believed that single crystals of this shape can be grown successfully.

From Eq. 7 it may be seen that for beta or alpha particles the pulse size is proportional to energy for *any* constant value of field strength (constant δ). It is generally desirable to use saturation field strength in order to obtain the largest possible signal. When gamma rays are counted with saturation field strength, the linear relation between pulse size and energy will not be preserved since the position of formation of a Compton electron (or photoelectron, pairs) is indeterminate and the pulse size is dependent on this position. Furthermore, the Compton electrons do not have a unique energy for a given gamma energy. The latter property means, of course, that a spread of pulse heights is inevitable even when counting monochromatic gamma rays. The spread in pulse height due to formation of Compton, etc., electrons at *different places* in the crystal can be minimized by making the freed secondary electrons all drift the same *small* distance in the crystal. A small field should be used in this case, resulting, unfortunately, in small signal pulses.

This manner of operating a crystal counter to detect gamma radiation was first pointed out by van Heerden (*6*), who has given the type of pulse distribution to be expected. The distribution curve can be calculated by assuming that: (1) all the ionization events produce an equal number (n_0) of secondary electrons, and (2) the ionization events are distributed homogeneously over the thickness of the crystal. With these assumptions the number of pulses (dN) lying in the range dQ at Q, where Q is the charge pulse, is

$$dN = \alpha d \frac{dQ/n_0 e}{1 - \frac{d}{\delta}\frac{Q}{n_0 e}} = \alpha d \frac{dq}{1 - \frac{d}{\delta} q} \quad (26)$$

where $q = Q/n_0 e$ and α is a constant which is proportional to the absorption coefficient of the ionizing radiation. The maximum pulse size is given by

$$\frac{Q_{max}}{n_0 e} = q_{max} = \frac{\delta}{d}\left(1 - e^{-\frac{d}{\delta}}\right) \quad (27)$$

and the distribution cuts off at this value of q_{max}. Corson and Wilson (*2*) have prepared interesting curves (Figures 16, 17, 18 of their article) which show pulse distributions of the type given by Eq. 26.

If the gamma-ray energies are high and the crystal large, it may be possible to use the crystal counter as a rough "gamma-ray spectrometer" since the pairs, produced in relatively large numbers in this case, will have the energy of the quantum except for a constant.

Search for Counting Materials

The published work on crystal counters makes little or no mention of the materials examined for the counting property. It appears to be unsafe to state that a given material "will not count" even after some tests on the material have been made. Some of the reasons for this caution are: (1) the material may neither be a single crystal nor made up of large crystal blocks, (2)

impurities may be present, (3) an amplifier may have been used which does not furnish the optimum signal-to-noise ratio—pulses may be present but are concealed by amplifier noise or by crystal conductivity, (4) particular conditions, e.g., annealing, low temperature, good electrode contact, good surfaces, absence of polarization, etc., may not be attainable under the specific conditions of the experiment, (5) unsuspected crystal imperfections and trapping centers may exist, (6) the "right" particle, to give the largest pulse, may not have been used (*47*), and (7) unknown causes, such as those responsible for making some diamonds gamma-ray counters and others not, may be present.

Jentschke (*9*) has remarked that the alkali halides and sulfur were observed not to count alpha particles. The writer has also examined,* with a 501 amplifier, one specimen of sulfur irradiated with beta particles of P^{32} without observing counting. Synthetic crystals of LiF, NaCl, KI(Tl), fluorite, Rochelle salt, and natural crystals of calcite and willemite have also been examined with gamma rays at different temperatures, without success. Particular attention was given to LiF because of the large value of its calculated mobility. Heat treatment and examination at different temperatures were attempted, all without success. D-C observations, with a 1.8 millicurie radium gamma source close to the specimen showed no current larger than 10^{-14} ampere although occasional, but not reproducible, currents larger than the above figure were noted. It is possible that polarization effects may have obscured the results in the latter case. These investigations were not pursued further. In any case, no individual pulses were ever observed in LiF samples, whether P^{32} beta particles or radium gamma rays were used.

* These remarks are made with reservation because of the reasons mentioned previously.

DISCUSSION OF RESULTS

Roughly speaking, when a crystal is shown to count, many of the observed counting phenomena find a natural explanation in terms of known facts. Some small discrepancies between theory and experiment have still to be investigated further, but no serious departure is known to exist. The energy per ion pair is quite low compared with the value for gases and it may be expected that a theoretical calculation will also show in detail how this comes about. The small energy gap between the filled band and conduction band in the silver halides makes an energy per ion pair of 7 volts or so quite reasonable. The value of 10 volts in diamond requires further explanation. Although most facts can be explained, it is still not known what makes a material a counter.

As observed previously, the alkali halide crystals do not act as counters although the earlier considerations connected with Table 1 do not indicate anything unusual about the mobilities in these materials.† This anomaly and the apparent absence of the counting property in many materials have puzzled the author for some time. He has proposed a tentative qualitative explanation (*45*) for the absence and presence of gamma-ray counting in diamond which suggests itself as a more general explanation for the absence of the counting behavior in most insulators, even those which are photoconductors.

This explanation may be put forth tentatively as due chiefly to the fact that most crystals, normally classified as single crystals, are in reality not single crystals at all. Since the early days of X-ray studies of crystals it has been found by Darwin, Moseley and

† F. Seitz has remarked to the writer that the presence of the acoustic modes of vibration in the alkali halides may lead to somewhat smaller values of l at low temperatures (77° K).

others (*61*, *62*, *63*, *64*) that most crystals, with rare exceptions, are mosaics. The meaning of this is that most crystals are composed of tiny crystalline (single crystal) blocks oriented almost parallel to each other but not exactly so. According to Darwin and Mark (*63*) the edge of a block, for the best typical mosaics, is of the order of one micron (10^{-4} cm). In some diamonds Mark has observed an angular misorientation of 2 minutes of arc, while in the most perfect NaCl sample studied the misorientation is of the order of 15 minutes of arc. Kirkpatrick and Ross (*64*) have also found from X-ray reflection studies that the width of reflected X-ray lines was of the order of 3.3 seconds of arc for calcite (perfect crystal gives 1.63 seconds of arc) while the best sample of NaCl gave a value of 87 seconds of arc (perfect crystal of NaCl gives 1.6 seconds of arc). These results indicate that NaCl crystals show much greater mosaicity than diamond crystals. Other samples of "good" NaCl are much more mosaic than those of Kirkpatrick and Ross—900 and 300 seconds of arc (*64*, *65*). P. P. Ewald (*66*) states that synthetic crystals of rock salt can be made with less mosaic structure than the sample of Kirkpatrick and Ross, although these are certainly not generally available.

It may be taken as well established, therefore, that NaCl, LiF (*67*), and probably other alkali halide crystals are mosaic crystals, whereas, on the other hand, it is possible to find excellent samples of diamond, calcite and ZnS which show tendencies of being perfectly regular single crystals. In most crystals, then, the freed secondary electrons are stopped at crystal boundaries and the average distance of motion is 10^{-4} cm or less. Under these conditions, a single pulse, due to say a 1.0-Mev beta particle, will be well under the noise level of most amplifiers in common use. However, it may be possible to observe integrated d-c current with a strong radioactive source in much the same way as photoconductivity currents are produced. In the case of diamond it appears that the ordinary Type I diamonds are microcrystalline mosaics and the Type II diamonds are constructed of laminations about 10 to 100 microns thick. The author therefore proposed (*45*) that Type II laminated diamonds should show better volume counting properties than the Type I diamonds, in agreement with the results of Friedman (*44*), *et al.* We might also expect that if the electric field lies parallel to the laminations larger pulses would be obtained than if the field is perpendicular to the laminations. (See below for further remarks on diamond crystal counters.)

It is clear that this explanation of the absence of the counting phenomenon does not furnish a positive criterion for a counting crystal. That is, a crystal which is not mosaic may not necessarily count because of other reasons such as traps, impurities, or cracks, although it is reasonable to expect that such imperfections are to be associated with mosaic rather than perfect crystals.* Other possible reasons for non-counting crystals may be a large value of the energy gap, ρ, an efficient mechanism within the crystal for transforming ion excitation energy into thermal vibrations of the lattice, or the possibility of "exciton" levels in the crystal (*32*).

The nature and number of traps in crystals are known only for a small number of materials and probably depend on the history and treatment of the particular specimen. It is to be expected that the counting phenomenon is a "structure sensitive" property of crystals and, in common with so many other structure sensitive properties, has no universal explanation. However, the study of materials which count is

* In this regard, it would be worth investigating good calcite samples for counting.

likely to increase the stock of knowledge of traps and trap mechanisms.

Mrs. K. Lonsdale (*68*) has questioned the validity of the explanation, proposed by the author, of gamma-ray counting or non-counting in Type II and Type I diamonds, and has specifically called attention to the fact that certain rare Type I diamonds are among the most perfect crystals known. This latter fact seems to be true, from the work of Lonsdale and others, although the author's remarks applied not to these rare specimens but to the average Type I diamond. One cannot make any positive predictions about these rare Type I diamonds for the reasons mentioned above, although it seems reasonable to assume that such diamonds would make good counters.

Mrs. Lonsdale has also proposed that electrons may slip along crystal boundaries in laminated diamonds. This cannot be a complete explanation of gamma-ray counting in Type II diamonds, as electrons must be produced in the body of the crystal before being able to reach the boundaries to travel along them. The possibility that a combined body and boundary motion may explain counting in diamond crystals has been suggested to the author by D. Bohm.

In discussions of diamond counters it is worthwhile to keep in mind that the distance secondary electrons may travel is probably of the order of 0.3 to 0.6 mm. These figures are based on McKay's (*19*) data on a Type II diamond which give $\nu T = 3\text{–}6 \times 10^{-6}$ cm²/volt and an assumed voltage gradient of 10^4 volts/cm.

C. V. Raman has kindly pointed out that a recent experiment by K. Achyuthan (*69*) on orientation effects in the photoconductivity of Type II diamond crystals shows that the conductivity is a maximum in the direction of the laminations. An effect of this kind was predicted by the author (*45*).

A discussion of alpha particle counting in various diamonds has been given by Ahearn (*47*) which shows that both Type I and Type II diamonds count alpha particles. Ahearn (*70*) has also reported, at the Rochester conference, local variations in the counting rate of diamonds when the face of a diamond was scanned by an alpha particle beam. The best counting in one particular diamond coincided with a flaw at the surface. No explanation has been given although it has been suggested that the flaw may have relieved local strains in the region. It may also be possible that electrons on the boundaries of the flaw are less tightly bound than in the body of the crystal, so that it is possible to form a large number of secondaries at the flaw. Ahearn's results are very similar to the local variations observed in photoconductivity by Achyuthan (*71*). It is apparent that the unraveling of the problem of the diamond counter awaits further experimental research. We see also that the more general question, "What properties determine whether a crystal will count?" is as yet unanswered.

Further experimental work is also required in the study of impurities within counters. Yamakawa and the author have prepared mixed crystals of NaCl-AgCl with as much as 15% by weight of NaCl. These samples behaved as counters, which suggests the possibility of preparing a neutron counter with a lithium impurity. The case of the mixed thallium halides (TlBr-TlI) also shows that mixed crystals can function as counters.

CONCLUDING REMARKS

Present Status

Linearity. The crystal counter is now only one of many different types of successful counters. Among the other useful counters are the Geiger-Müller counter, the ionization chamber, the proportional counter, the electron

multiplier counter, the scintillation counter and the new parallel plate counter (*72*). The ionization chamber, proportional counter and crystal counter are linear devices, that is, counters in which the pulse height is proportional to the energy of the incident particle. In order that such counters be linear the particle must lose all its energy within the counter. In the case of ionization chambers and proportional counters it is difficult to make an energetic beta particle lose all its energy in the gas of the counter unless a high pressure is used, with its attendant difficulties. With a crystal detector, on the other hand, it is a relatively easy matter to stop a beta particle of energy up to 10 Mev in a small sample. Thus, linearity of behavior, even for energetic particles, is one of the strong arguments for use of a crystal counter.

Stopping power. A major use of a crystal counter is likely to arise also in its application to detection of gamma rays. The stopping power of a crystal is enormously higher than that of a gas and it is this fact that has led to the great interest in crystal and scintillation counters. It has been mentioned that small crystals may give efficiencies of the order of 10–15 % for radium gamma rays. A large crystal specimen may be used to stop a 100-Mev gamma ray and all the products of its interaction with matter (Compton electrons, pairs, Bremsstrahlung, X-rays). The high density (7.5 gm/cm³) of TlBr-TlI suggests further investigation of its properties along these lines. It is conceivable, in fact, that large crystals may be devised to behave as gamma-ray energy sensitive devices, the pulse size giving directly the energy of the gamma ray. In view of the extreme difficulty of measuring energies of hard gamma rays by other methods (*73*, *74*), it seems well worthwhile to investigate the crystal detector as a gamma-ray energy spectrometer. In this connection it seems reasonable to employ a long flat box-like crystal with electrodes on its wide faces with the gamma-ray beam entering parallel to the long direction. Since the energy is high for hard gamma rays, the voltage gradient will preferably be small. In this case the position of conversion of the gamma ray to Compton electrons and pairs will be unimportant unless extremely close to the electrodes, as the secondary electrons will drift only a short distance in the crystal before being trapped. Large single crystals of AgCl have been grown commercially so that the remaining problem will concern the heat treatment or annealing of such crystals. In this writer's opinion such a problem can be solved.

At the present time the scintillation counter vies with the crystal counter for high energy applications since fluorescent materials of high density have been discovered recently [$CaWO_4$ (*75*) density 6.06 gm/cc and NaI (*76*) density 3.67]. However, a problem still remaining is the collection of all the light from such a large crystal.

Resolving time. In the matter of resolving time the crystal counter is in an intermediate position. While not as fast as the organic scintillation counters, phenanthrene and stilbene (0.02 microsecond), the resolving times, which are either transit times or trap capture times, are of the order of a few hundredths to one or two microseconds. In diamond the resolving time is of the order 0.04 microsecond. The crystal counter is considerably faster than the Geiger-Müller counter, both in respect to duration of the pulse and initial delay of the pulse, the latter being absent in the crystal counter. Ionization chambers and proportional counters are also relatively slow (3–10 microseconds). The crystal and scintillation counters both offer the attractive possibility of measuring the time interval between two events which occur in the same

counter material, even though the time interval is of the order of, or less than, the duration of the individual pulses. It must be remarked that amplifier noise caused by wideband operation places a limit on the speed of the crystal counter unless the pulse corresponds to a large energy. Such a restriction is not binding on the scintillation counter.

Lifetime. The useful lifetime without treatment of a crystal counter depends on the application. For penetrating radiation the field may be periodically reversed to avoid polarization effects and the lifetime would appear to be very great, perhaps unlimited. For beta particles which penetrate only a short distance within the crystal (AgCl, AgBr) the lifetime before reheating is probably of the order of a few million counts for a small crystal. Reheating and recooling allows the crystal to be used indefinitely. However, there is no question that the polarization effects place serious handicaps on the crystal counter. For, as the crystal polarizes, the pulse size may vary during an experiment. Efforts made to release frozen charges at a slow rate are definitely recommended. The scintillation counter would appear to have very long life.

Lowest detectable energy. This quantity is probably the energy equivalent of 100 or 200 electrons with present-day tubes (6AK5) and therefore, in principle, about one or two electron kilovolts. However, this low energy places a limit on the speed of counting at about 160 counts per second. The lowest detectable energy for a scintillation counter is not known but is probably less than five kilovolts. The Geiger-Müller counter can detect energies less than one kilovolt, the only limitation here being the window thickness. Windows are not required in crystal and scintillation counters.

Limitations on experimental possibilities. It is in this category that the low temperature crystal counter suffers greatly with respect to other counters. This may be a temporary situation which can be ameliorated by discovery of a new efficient room temperature counting material. However, at present, when one desires to make use of the large amount of material in a solid by employing a large crystal, he is limited to the silver or thallium halides which require low temperatures and annealing procedures. Both requirements force one to use bulky, massive and complicated cooling and vacuum systems. In some experiments, for example, scattering problems involving gamma rays, the dewar and vacuum chamber walls must be present and can add seriously to background or spurious counts.

In the case of small detectors it seems that it will not be possible to improve greatly on diamond or CdS. Diamond or CdS counters may be superior to scintillation counters in this respect as they merely require two connecting wires, whereas the fluorescent material requires a photomultiplier in a dark box or a light-piping scheme.

FUTURE POSSIBILITIES

It will appear to the reader that the crystal counter field has been barely opened by the past and current efforts. However, the quite likely discovery or preparation of a new counting material may alter matters considerably. There is certainly more than just a hope that some known or new material may be perfected to serve as a room tempera-

Note added in proof: Two papers (*78,79*) on CdS have recently appeared. The paper by Frerichs and Siegert deals with gamma-ray irradiation and trap configurations. Information on traps has been obtained by studies at different temperatures. The work of Goldsmith and Lark-Horovitz concerns pulse size and pulse rise times in CdS. Quite large pulses (12 mv for Po alpha particles and 6 mv for RaE beta particles) were observed by the latter authors, indicating a low energy per ion pair. Pulse size distributions are also given in this paper. Pensak (*80*) has observed electron-bombardment-induced conductivity in quartz and other substances.

ture counter, or that some new tube or advance in the electronic art may be made which will allow amplification with a lower noise level. In crystal counter applications the crystal does not appear to have noise comparable with the noise introduced in bringing the signal to a higher power level.* While much interest has been drawn from the crystal counter to the scintillation counter by the rapid and spectacular advances in the latter field, there is still a great deal to be learned about the solid conduction-type counter both in respect to obtaining fundamental knowledge of the crystalline state and in regard to its uses as a new tool for studying penetrating radiation. The remarkable results of Kallmann and Warminsky with CdS may encourage research in both these directions.

* * *

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* E. S. Rittner (*77*) has demonstrated bombardment conductivity in the semiconductor selenium. It may not be possible to observe single conductivity pulses due to an ionizing particle in a semiconductor because of the electronic conductivity. However, the possibility is so attractive that ways and means may yet be found to demonstrate such an effect.

COSMIC RAYS—II

A review of present knowledge in a field in which a vast program of research is taking place, together with a report on the status of this research leading to a better understanding of the fundamental properties of the elementary particles

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The Hard Component

FOR MANY YEARS the nature of the hard or penetrating component constituted one of the most puzzling problems in cosmic ray physics. The solution to this problem was found in the discovery of a new elementary particle which received the name of *mesotron*.

The hard component consists of about 55% positive and 45% negative particles. The main difficulty arose from the fact that it is impossible to reconcile the properties of these particles either with electrons (positrons) or with particles of protonic mass, which are the only known charge-carrying particles in atomic physics.

That the particles are not electrons can be seen from the fact that they are much more penetrating than electrons and are not shower producing. In early times there was some doubt whether the quantum theory of radiation could be applied to particles of such high energy. Experimentally, however, the validity of radiation theory could be very well established up to the highest energies available with cosmic ray electrons.

On the other hand, the particles in the hard component cannot be protons since the measurement of the curvature in a magnetic field of the slower particles, together with their specific ionization, gives values which do not agree with the corresponding data for protons, quite apart from the fact that nearly equal numbers of positive and negative particles are observed and the negative proton is not known to exist.

In short, these two experimental facts tell us, therefore, that the mass of the hard component particles is much larger than the electron mass, since their radiation loss is much less than that of electrons (the radiation loss of a particle is inversely proportional to the square of the mass), and considerably less than the proton, since tracks of equal specific ionization are much more strongly curved in a magnetic field for the hard component particles.

A large amount of additional evidence was gradually accumulated which corroborated these findings and led finally to the discovery of the mesotron. The nature of the mesotron is not yet fully understood. A great deal of research is in progress and new results bearing on this question are being published in rapid succession. We shall here briefly summarize the present state of our knowledge of the nature of the mesotron.

The most recent and probably the best mass determination is due to W. B. Fretter (*29*) who finds a value of 202 electron masses with a statistical error

of ± 5 e.m. for the cosmic ray mesotrons at sea level. There is no indication of more than one mesotron mass. Substantially the same conclusion is reached by Hughes (*30*) who analyzed some fifty mass determinations by many different methods and investigators.

Recently (*31*), however, conclusive evidence has been presented for the existence of a mesotron in the hard component of the cosmic radiation with a mass larger than the ordinary mesotron mass. Such particles were first discovered in photographic plates exposed to cosmic radiation at higher altitudes. This important result is corroborated by the reports from Berkeley according to which mesotrons can be produced artificially with α-particles of 380 Mev energy. The mesotrons observed could clearly be divided into two groups, one of mass about 216, and the other of about 286 e.m.

The second relatively well-established fact about the mesotron is its radioactivity. Mesotrons decay spontaneously probably into an electron and a neutrino. The decay time for the ordinary mesotron (we refer to the mesotron of mass 200 e.m. as the "ordinary mesotron," since it is apparently the most abundant in the hard component of the cosmic rays) is found to be 2.2×10^{-6} sec.

The most direct evidence of the decay process is cloud chamber pictures (*32*) which show tracks of mesotrons which get stopped inside the chamber and show a decay electron starting from the end of the mesotron track. The two kinds of tracks are easily distinguishable on account of their difference in specific ionization.

Another evidence is the anomalous absorption of mesotrons in air. Mesotrons in air show larger absorption than in the same amount of a denser material. The difference can be accounted for with the decay of the mesotron. Since a mesotron of given momentum spends a longer time traversing a certain amount of air than traversing the same amount of a denser material, the probability for its decay in air is larger than in the same amount of a denser absorber. Consequently fewer mesotrons will be recorded behind air than behind the denser absorber.

The most precise determination of the lifetime of mesotrons, however, were made with a delayed counter arrangement which in principle may look as indicated in Fig. 8. The three counters *A* are in coincidence and separated by about 10 cm of lead. Thus they register only mesotrons which enter in the vertical direction. The counters in tray *B* are in coincidence but in anticoincidence with the system *A*. Thus only mesotrons will be registered which are stopped in the absorber *b*. The counters in tray *C* are all in parallel and in delayed coincidence with *A*. The delay time may be adjusted to vary over several microseconds. Thus the system as a whole registers only mesotrons which are stopped in *b* and are followed by the emission of an ionizing particle several microseconds later. From the number of particles which are ejected for various time delays one can obtain a decay curve for the mesotron.

When this experiment was carried out with a heavy absorber *b*, it was found that only about half of the mesotrons which are stopped in *b* are followed

EDITOR'S NOTE. In Part I of this paper, published last month, the following subjects were discussed: Discovery and Experimental Methods, Altitude Dependence of Cosmic Rays, Components of Cosmic Rays, Geomagnetic Effects, and the Soft Component. The numbering of sections, figures, and references in this second half of the paper is continued from the first half.

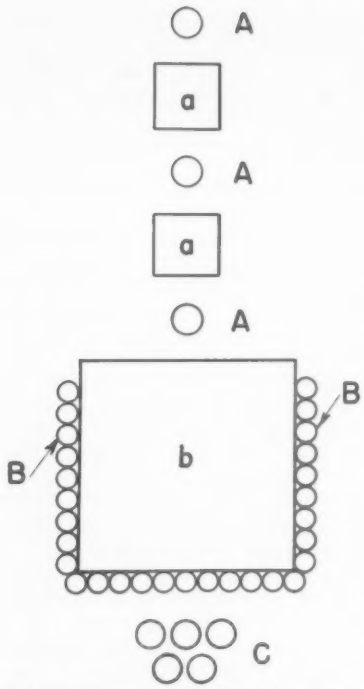

FIG. 8. Counter arrangement for measuring the lifetime of mesotrons

by a decay electron. This result was expected on the basis of Yukawa's theory of the mesotrons. As early as 1935, Yukawa (*33*) postulated the existence of a new elementary particle on purely theoretical grounds. This particle was to be intimately associated with the nuclear forces. Yukawa's theory predicted that the particles would have a mass of about 200 mass units and that they would be radioactive. When the cosmic-ray mesotrons were discovered with just these properties there seemed no doubt that Yukawa's particles were real and were here actually observed. According to this theory these particles would interact strongly with the nucleons (neutrons and protons) and, according to the calculations of Tomonaga and Araki (*34*), negative mesotrons have a much larger probability of getting captured in a nucleon than undergoing a decay. Thus only the positive mesotrons should be observed with a delayed decay electron.

At first the mesotron theory of Yukawa looked very promising. However, there were several details which did not agree very well with the experimental facts. In the first place, the radioactivity predicted by the mesotron led to a lifetime of about 10^{-8} sec, which is roughly one hundred times smaller than the actually observed lifetime. Next, the actually observed mass of the cosmic-ray mesotron does not fit very well the mass value deduced from the interpretations of proton-proton scattering. Hoisington, Share and Breit find a mass value of 326 in good agreement with the scattering data but 200 e.m. quite outside the experimental error. Finally, the experiments with the delayed counter coincidences just discussed showed (*35*) that in a light absorber *b* (Fig. 8) such as Be or C, all the mesotrons decay, whereas in a heavier one such as Fe or Pb only the positive mesotrons decay. A quantitative comparison of this result with the result deduced from the Yukawa theory reveals (*36*) that such a behavior could be expected only if the strength of the interaction were of the order of magnitude 10^7 times smaller than is needed for the theory of nuclear forces.

This state of affairs must be contrasted with another body of information relating to the origin of the mesotrons in the atmosphere. It is almost certain that no mesotrons enter the atmosphere from the outside. This may be concluded most easily from the fact that they are radioactive with a lifetime of only a few microseconds and therefore could not possibly

survive the long time needed to reach the surface of the earth from interstellar space. Another reason is that the mesotrons observed at sea level carry positive and negative charge in almost equal amount. If they were primaries there should be practically no east-west effect at sea level in disagreement with the observations. A third reason is found in the fact that according to Blackett the average energy of the mesotrons at sea level is only about 3 Bev. The total energy loss in the atmosphere as estimated from the specific ionization amounts to 2 Bev. If the mesotrons were primaries, their average energy at the top of the atmosphere would therefore be 5 Bev. But this is impossible since it follows from the latitude effect of the hard component that 90% of the primaries responsible for the hard component at sea level must have energies larger than 15 Bev. This argument shows in addition that, in the average, each primary produces from 3 to 5 mesotrons which share the initial energy of the primaries.

It follows therefore that the mesotrons are secondaries produced by some other kind of primary radiation. Since there is strong evidence that the primaries of the hard component are protons (see below), we must expect that protons upon entering the atmosphere produce the mesotrons in a collision process with the nuclei of the air atoms. From the fact that very few fast protons are observed even at very high altitude it follows that the cross section for such processes must be very large.

A quantitative theoretical study of the production of mesotrons by primary protons was made by Hamilton, Heitler and Peng (*37*). In this theory the protons are supposed to interact with mesotrons with a strength determined essentially by the nuclear forces. Two types of mesotrons are considered, the pseudoscalar (spin 0) and the vector (spin 1) mesotrons. The vector mesotron is assumed to decay with a lifetime of approximately 10^{-8} sec (the time required in Yukawa's theory of the β-decay) whereas the pseudoscalar mesotron is supposed to have the observed lifetime of the ordinary cosmic ray mesotron. The theory predicts that the primary protons with energies larger than 1 Bev will radiate mesotrons at such a rapid rate that they lose most of their energy in an atmospheric layer of about 50 gm cm^{-2}. The vector mesotrons then disintegrate rapidly into electrons and neutrinos, thus giving rise to the soft component at high altitude. The longer-lived pseudoscalar mesotrons, on the other hand, are the ones which reach the surface of the earth and form the hard component at lower altitude. Qualitatively the theory gives the right altitude dependence of the two components. A quantitative comparison with the experiments is meaningless for several reasons. In the first place, mesotron production in this theory is considered as a single elementary process such that in each process exactly one mesotron is produced. Lately, however, experimental evidence has been accumulated which shows that mesotrons can be produced in bursts of from 5 to 10 mesotrons in a single process. Furthermore, the pseudoscalar mesotron is assumed to interact strongly with the protons, as it follows from the mesotron theory of nuclear forces. But this is no longer possible in the light of the newer experiments on the decay of negative mesotrons in absorbers just discussed above. In addition, it has been observed recently that the heavy mesotrons may decay into the lighter ones, a process which was not assumed in the mesotron theory of nuclear forces underlying the calculations of Hamilton, Heitler and Peng. Finally, it has been shown by Schein and collaborators (*38*) that mesotrons are also produced by a non-ionizing radiation.

It is thus clear that a more complete theory is needed for giving a quantitative description of the distribution of the components at various altitudes as well as the geomagnetic effects. There seems no doubt, however, that mesotrons are produced in the higher atmosphere at an exceedingly rapid rate. On the other hand, the mesotrons at sea level interact only very weakly with the nucleons. Therefore, mesotrons produced by the primaries are different from mesotrons observed at sea level.

As a working hypothesis one may assume that the heavy mesotrons interact strongly with the nucleons and are fast decaying (lifetime about 9×10^{-9} sec). The ordinary mesotrons are decay products of the heavy mesotrons and interact only very weakly with the nucleons.

A partial check on this hypothesis can be found by a direct measurement of the spin of the mesotron. This seems very difficult since the mesotrons have such a short lifetime. It has however been possible to make some assertions about the spin of the mesotrons in cosmic rays by measuring certain electromagnetic effects due to the spin. The underlying theory is based on the calculation of the emitted electromagnetic radiation for a close collision of an electron with an atomic nucleus (*39*). It turns out that at sea level the large bursts of ionization in a shielded ionization chamber are largely due to this radiation. Moreover, the calculation shows that the dependence of the frequency of occurrence for such bursts on the size of the bursts is very sensitive to the spin of the mesotrons which produce the radiation. The size-frequency curve measured by Lapp (*40*) over a period of five years taken at sea level can be made to agree either with spin 0 or ½ but definitely excludes the value 1. We conclude therefore that the mesotrons at sea level have spin smaller than 1.

On the other hand the same measurements at higher altitudes give a curve which lies between the curves for spin ½ and 1. Thus this experimental result is in agreement with the theory that the cosmic-ray mesotrons at higher altitude contain a mixture of two different kinds of mesotrons, one of spin 1 and the other of spin 0 or ½. The spin 1 mesotron would presumably have to be identified with the heavier mesotron recently observed on photographic plates.

We turn now to the geomagnetic effects of the hard component. The latitude effect at sea level is essentially that of the total radiation and amounts thus to about 9%. At higher altitude, the latitude effect, as measured by Gill, Schein and Yngve (*41*) and by Swann and Morris (*42*), was found to be 30% at 270 gm cm^{-2} atmospheric depth. In contrast with this, Bhabha (*43*) and co-workers infer from a comparison of their measurements with those of Schein and co-workers that at the same altitude the effect amounts to only 18%. This value may, however, be subject to large error because it was obtained by a comparison of experiments which may have a different efficiency for recording the hard component.

The east-west effect at low altitude is essentially that of the total radiation and amounts to about 15% (positive) at $z = 45°$ and $\lambda = 0°$. It decreases with increasing value of λ. Finally the east-west effect of the hard component at higher altitude was recently measured by Yngve, Schein and Kraybill (*44*) and was found to be 45% for $z = 45°$, $\lambda = 27°$ and depth of about 240 gm cm^{-2}. It is expected to be even larger at lower geomagnetic latitude and higher altitude.

The comparison of these high positive values for the east-west asymmetry with the value obtained from the Lemaitre-Vallarta theory reveal that most of the primaries of the hard component must be positively charged.

Penetrating Showers and Stars

The cascade showers which were discussed in Part I are not the only kind of showers which occur in cosmic rays. For a long time various workers occasionally obtained cloud chamber tracks of showers which could penetrate several radiation units of lead without undergoing the usual multiplication which is so typical for the ordinary electron showers. A study of such showers revealed that they probably consisted mostly of protons. Occasionally they may occur with nuclear fragments. Direct observations of nuclear disruptions which break up the nucleus into several parts were first observed by Blau and Wambacher (*45*) on photographic plates. Several heavily ionizing tracks occurred with a common origin, giving the appearance of a star.

The frequency of occurrence of both the penetrating showers and the stars increases rapidly with increasing altitude. The increase is approximately exponential: $e^{-x/L}$, with a value of $L \simeq 125$ gm^{-2} and is about at the same rate as the increase of the soft component. From this fact it has been concluded until recently that stars are produced by photons of the soft components. However it is now fairly well established that neither the nuclear disruptions nor the penetrating showers are produced by the soft component.

Cloud chamber pictures which were made under lead show stars and penetrating showers which are not accompanied by electrons. This would be impossible if these events had been initiated by a photon or electron since they would be accompanied by a shower. Similar experiments were made with counter arrangements which could detect the accompanying showers.

Various experiments show that neither the penetrating showers nor nuclear disruptions show any appreciable reduction behind thick layers of lead. For the latter process, for instance, a lead absorber of 30 cm showed a decrease in the frequency of only 30%, which is quite incompatible with the assumption that a soft component particle originates the process. Both the altitude variation and the behavior behind absorbers are in agreement with the assumption that the penetrating showers and the stars are produced by fast protons and neutrons. It is not impossible, however, that, in addition, heavy mesotrons or neutral mesotrons may be responsible for these events.

It has also been found that some of the penetrating showers in air are often accompanied by large air showers. Cloud chamber pictures also often show the simultaneous occurrence of soft showers with the penetrating showers. At present the exact mechanism of the penetrating showers is not well enough understood to interpret this fact with any degree of certainty.

The Primaries and the Genesis of Cosmic Rays

In the previous sections we have presented most of the experimental facts which have any bearing on the question of the nature of the primaries and the subsequent transformations which these primaries undergo in the atmosphere. A complete theory of cosmic rays in the atmosphere would involve a calculation of the known component structure, their intensity distribution, their variation with height and their geomagnetic effects in terms of the elementary cross-sections for the individual transformation processes. There are as yet too many uncertainties involved to give such a theory at the present stage of cosmic ray research. Nevertheless, certain qualitative statements can be made at this time on the basis of the known phenomena with great certainty.

We begin with a discussion of the nature of the primaries. The numerous experiments on the geomagnetic effects of the hard component establish

almost with certainty the fact that the primaries of the hard component are protons. The question remains yet whether there are other primaries besides the protons. In the first place, the neutrons are not likely to be among the primaries for various reasons. The free neutrons are unstable, with a probable lifetime not larger than a few hours. This is sufficient to rule them out as primaries. Moreover, if they were responsible for an appreciable fraction of the hard component, the geomagnetic effects would be quite different from the observed effects. On the other hand, the important experiments by Schein, Jesse and Wollan (*26*)* and seven years later by Hulsizer and Ross (*27*)* rule out photons or electrons as primaries. We have already discussed the reasons why mesotrons are out of the question as primary particles. With this we have exhausted the list of known elementary particles. There still remains the possibility that positive ions of heavy elements occur among the primaries, either partially or wholly stripped of their surrounding electrons. Such ions have recently been found with the photographic plate method, and their relative frequency was found to be such as to constitute about 0.1% of the total incoming primaries. The presence of these ions in the primaries may have important consequences for the speculation about the origin of cosmic rays, but for the transition effects in the atmosphere they have only little influence.

Unless we want to invent new particles which are not yet observed, we should try to examine the hypothesis that the total primary radiation consists of protons (with the exception of the above-mentioned heavy ions).

The crucial experiment is that by Johnson and Barry (*25*)* on the east-west effect of the total radiation at very high altitudes (30–60 gm cm^{-2}). It was pointed out by these authors that the small east-west effect of the total radiation together with the known latitude effect would require that the charges of the primaries which are responsible for the soft component at high altitude would have to be negative and positive. Since this experiment was performed, the east-west effect of the hard component at high altitude has also been measured and reveals a very large positive value which would make the east-west effect of the soft component at high altitude possibly even negative. To this, one must remark that the soft component at high altitude contains a large number of low energy electrons (very few penetrate 4 cm of lead, which corresponds to a critical energy of about 600 Mev). If they have their origin in a decay process of fast-decaying mesotrons of low momentum, or are the by-products of penetrating showers, they would be expected to have undergone a considerable change in direction from the original direction of the primaries. Thus a smaller east-west effect than that of their positive primaries would be expected. To decide this question, an experiment which measures directly the east-west effect of only the soft component at very high altitude (not the total radiation) should be made. Then the effect of the above-mentioned angular spread of the soft component should be calculated and the result compared with such an experiment. Before this is done we feel that the evidence is not sufficient to rule out the hypothesis that all the primaries are protons.†

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* Reference in Part I.

† The experiment mentioned here has now been performed by W. C. Barber *Phys. Rev.* **75**, 590 (1949). He finds the asymmetry for the soft component much larger than Johnson and Barry (*25*) and not in disagreement with the assumption that all primaries are positively charged. Although the east-west effect of the soft component is still somewhat too low, the author feels that the deficiency in the asymmetry can probably be explained by the change in direction which the secondaries have undergone in the production process.

To avoid the above-mentioned difficulty with the east-west effect of the soft component at high altitude, Arley (*46*) has postulated the existence of negative protons in the primaries. Such negative protons are possible in Dirac's theory of holes as applied to the ordinary protons. They have never been observed in any experiment, however, and their existence is rather hypothetical since the use of Dirac's theory for a proton is not necessarily correct in view of the anomalous value of the magnetic moment.

Although it removes the difficulty connected with the Johnson-Barry experiment, this hypothesis is nevertheless untenable. There are two main reasons for this. In the first place, most of the energy of the incoming negative protons would have to be in the field-sensitive region of the radiation, that is, around 2–15 Bev. According to Arley they would be annihilated at the very top of the atmosphere under the emission of two photons. These photons would then be the parents of a beginning cascade process which would furnish the soft component near the top of the atmosphere. Below about one radiation unit of air there would still be a large number of electrons with energies larger than 1 Bev. However, Schein (*26*)* and collaborators have found that practically all the electrons at one radiation unit below the top of the atmosphere have energies less than 1 Bev.

The second reason against this theory is that the cross section for the annihilation process as calculated from the Dirac theory is smaller by a factor 10^7 than the value needed to explain the large number of secondary electrons at high altitude.

Therefore, we conclude that the assumption of protons as primaries is in best all-around agreement with the known experimental facts on cosmic rays.

It remains to be shown how this proton hypothesis accounts for the known component structure of the cosmic radiation at various depths in the atmosphere.

In the first place we need a mechanism by which the primary protons lose energy very rapidly and produce the intense mesotron radiation at very high altitude. The experiments at high altitudes indicate that mesotrons are probably produced in multiples ranging perhaps from 5 to 10 for each individual process depending probably on the energy of the primaries. If the Berkeley experiments are considered a safe indicator of the mode of mesotron production we would expect that the mesotrons thus produced are of the short-lived heavier type which decay in about 10^{-8} sec into the longer-lived mesotron of mass 200 e.m. usually observed at sea level.

Primary protons may also start showers either by bremsstrahlung or pair creations or by as yet unknown processes giving rise to the larger air showers. As was already indicated, these showers, however, cannot be considered as the main part of the soft component since experimentally only a few per cent of the component at high altitudes can have energies larger than 1 Bev. The main contributions to the soft component at high altitude must come from two sources. First is the decay of the ordinary mesotron. This process is almost certainly the main source of the soft component near sea level. At higher altitude it does not seem enough. There must exist still another as yet little understood process, which furnishes the main contribution to the soft component at high altitude. It must be a relatively effective process which is initiated probably by very high-energy particles but furnishes electrons of considerably

* Reference in Part I.

lower energy (less than 10^9 ev). One would expect thus some kind of multiple process whereby a large number of electrons are produced in one single process.

As one goes farther down into the atmosphere the composition of the components changes gradually. The heavier mesotrons are eventually replaced by the lighter (ordinary) mesotrons and some of the latter decay and others collide with electrons in the air molecules thus contributing more to the soft component. The soft component on the other hand first multiplies and then gets absorbed but is partly replenished by the hard component.

This qualitative picture of the genesis of cosmic rays in the atmosphere shows that a quantitative theory would be quite complicated. It is very difficult at the present stage of knowledge to go beyond a qualitative description especially for the effects at high altitude since the fundamental processes involved are practically unknown.

In conclusion of this section we should mention that there may be neutral mesotrons present which in turn may decay into a pair of charged particles. Neutral mesotrons have not yet been observed, but their existence is postulated in the recent forms of the theories of the nuclear forces. A quantitative theory has little chance of success without some definite information on the role of neutral mesotrons in cosmic rays.

Speculations on the Origin of the Primaries

A general review paper on cosmic rays would not be complete without a discussion of the speculations which have been made so far on the origin of the primaries. At the present time a complete theory of the origin of the primaries does not exist.

Any satisfactory theory must explain the following properties of the primaries which were discussed in this review and which we summarize here once more for convenience:

1) The majority of the primaries are protons. At least one in one thousand primaries is a heavy positive ion with nuclear charges ranging between those of carbon and iron. It is possible and in fact very likely that α-particles enter as primaries also, but apparently this is not known yet.* No primary electrons are observed.
2) The total directional intensity of the primaries is 0.12 particles cm^{-2} sec^{-1} $sterad^{-1}$. They are nearly isotropically distributed (anisotropy less than 0.05%).
3) The primaries have an integral energy spectrum of the form

$$F(E) = 0.05 \left(\frac{10^{10}\,\text{ev}}{E}\right)^{1.6} \text{sec}^{-1}\text{cm}^{-2}$$

for energies $E \gtrsim 6 \times 10^9$ ev.† Primary energies up to 10^{14} ev are established with certainty and strong indications point to energies as high as 10^{17} ev.
4) The periodic variations of the primary intensity with the periods of the solar system are exceedingly small and can be adequately explained as secondary effects primarily due to the magnetic field of the sun.

The following conclusions seem rather obvious and are the starting point of almost all attempts to make a theory of cosmic rays:

a) Cosmic rays are apparently really

* Since this article was written several workers have detected α-particles in the primaries both with the Wilson cloud chamber and the photographic plate. The abundance of α-particles is estimated to be about 20% of the total radiation.

† This energy spectrum refers to the total radiation regardless of the composition of the primaries. The important question whether the heavier nuclei which are present in the primaries follow the same energy spectrum is at present under investigation but not yet decided. Preliminary indications seem to favor the view that the energy spectrum of heavier nuclei is different from that of the protons.

cosmic in origin. That is, they have their origin probably far outside the solar system. Furthermore, a normal star like the sun is apparently not a source of cosmic rays. Otherwise we should expect the sun itself to be a strong source which would give rise to strong periodic variations.

b) The simple energy spectrum, which seems to be valid over several powers of ten for the energy, points to a single process of the total radiation rather than a superposition from basically different processes.

c) The almost complete isotropy favors sources which are either fairly uniformly distributed or in which the magnetic fields are sufficiently strong to change the directions many times between the source and the observation point.

We shall omit the discussion of the theories which are in obvious disagreement with any of the facss about the primaries summarized above. A first sifting according to this principle leaves us essentially with two types of speculations which merit a more careful examination:

a) The origin of cosmîc rays is seen in some kind of annihilation process.

b) Cosmic-ray energies originate in an acceleration process in electric fields.

Before discussing some specific theories of this sort it is desirable to refer to some difficulties which are encountered by any theory.

If the radiation originates inside the galactic system then such radiation is constantly streaming out of that system into intergalactic space through the boundary of the galaxy. If we assume that the density at the earth is typical for cosmic rays all over the galaxy, a simple calculation shows that the net flux of energy out of the galaxy is about 10^{42} erg sec^{-1}. This seems an enormous amount of energy since it is comparable in amount to the rotational energy of an average star. Since there are about 8×10^{10} stars in the galaxy the total energy emitted during the lifetime of the galaxy ($\sim 10^{10}$ years) would be more than 10^7 times the rotational energy of all the stars.

This is of course not a conclusive reason against an intragalactic origin. There are several possibilities to get around such consequences. In the first place we may assume that cosmic radiation exists not only inside the galaxy but also in intergalactic space and that therefore the net flux is very small if not altogether zero. In the second place, if there exists an axial magnetic field as small as 10^{-11} gauss inside the galaxy, the cosmic ray particles are practically trapped inside the galaxy. For a 20-Bev proton in such a field has a radius of about three light years, which is entirely negligible compared to the major axis of about 35,000 light years of the galaxy.

To the first of these alternatives we must remark, that a practically uniform distribution in intergalactic space is subject to a partial experimental check. Such a distribution would be expected to be isotropic in a coordinate system which is at rest with respect to the center of gravity of the nearest galaxies. It would not be isotropic for a point such as the sun which is partaking in the general rotation of the galaxy. An experimental check of this effect has been attempted. The results seem to favor the absence of such an anisotropy although the evidence is rather uncertain (*47*).

The second alternative has been discussed by various authors (*48*), and from their discussions it seems likely that such fields of the order of magnitude 10^{-12} to 10^{-8} gauss may very well exist inside the galaxy. It may be worth pointing out that the galaxy is

then practically a closed system for cosmic ray particles, since not only are they trapped in the galaxy but no particles can enter from the outside either.

Another difficulty which should be discussed is the propagation of cosmic rays. Cosmic ray particles which are charged produce a space charge and may also produce magnetic fields, which in turn may limit the propagation of such charged particles through space. For instance it has been remarked by Swann (*42*) that if cosmic ray particles are charged and uniformly distributed they would have a space charge which would produce a potential difference between the center and the periphery of a sphere of radius R in light years given by

$$10^{18}\ R^2 \text{ volts}$$

This result may not mean as much as it seems, since it is now known that interstellar matter is ionized and must be regarded as a good conductor. Thus such space charges cannot maintain themselves for any reasonable length of time. Another kind of limitation pointed out by Alfvén (*50*) seems much more serious. Alfvén remarks that the transport of charged particles over galactic distances constitutes a current the magnetic field of which imposes strong limitations on this current. As a consequence, Alfvén finds that cosmic ray intensity may very well not be uniform in intragalactic space. Furthermore, he sees in that a reason to believe that the source of the cosmic rays observed on the earth may be less than 1000 light years away. It is difficult to appraise these qualitative conclusions. They may at best be considered an indication that a more precise and quantitative theory is needed and worth while for the propagation of charged cosmic ray particles in space.

Turning now to the discussion of the actual hypothesis regarding the origin of cosmic rays, we shall discuss first the theory which sees the origin of cosmic ray energies in some kind of annihilation process, whereby an atom of mass M is annihilated and then transmits its total rest energy Mc^2 to two or more outgoing particles. Theories of this sort were discussed very early in the history of cosmic ray research. Of course they have undergone considerable change and adaptation as cosmic ray research revealed more and more of the nature of the primaries. In spite of their rather sensational character there are still some prominent physicists who take it seriously.

The starting point of the annihilation theory is the observation that the rest energies of the most abundant elements (with the exception of H) in the universe, namely He, Si, C, O, N, are not very different from the average energy of 10 to 20 Bev of the primary cosmic ray particles.

Half the rest energies of these atoms (which would be the energy taken up by one of a pair of particles created in the process) is approximately given in the following table together with the approximate geomagnetic latitude in the western hemisphere where they would first enter vertically:

	Energy in Bev	*Geomagnetic latitude*
He	1.88	54°
C	5.6	40.7°
N	6.5	40°
O	7.5	32.8°
Si	13.2	0°

It is clear from the beginning that this fact may not be more than a coincidence and may therefore have nothing to do with the true nature of the cosmic rays. There is no way of deciding such a question unless the theory can be correlated somehow with other experimental facts and can give an internally consistent picture of the cosmic ray phenomena.

It has been pointed out by Millikan (*49*) and collaborators that such an experimental verification of the atom annihilation hypothesis is, in principle, possible. For if the energies of the incoming cosmic rays are discrete with the values indicated in the table, then the vertical intensity curve as a function of latitude should have a plateau structure. The intensity should show a sudden change always at the critical geomagnetic latitudes and should be nearly constant in between. For this reason Millikan and co-workers have carried out a series of very careful measurements of the vertical intensity as a function of the geomagnetic latitude.

Before entering into a discussion of this experimental evidence, we shall take up some of the difficulties connected with the atom annihilation hypothesis. First, it may be worth while to look somewhat more closely at such an annihilation process. In the discussions on this hypothetical process, undue prominence has been given to the conservation of energy and not enough to the other conservation laws, namely that of linear momentum, angular momentum, and charge. The linear momentum conservation is relatively easy to satisfy by postulating a pair formation. It is not quite so easy, however, to satisfy the conservation laws of angular momentum and of charge. Let us look for instance at the charge conservation. In the original forms of the theory a two-quanta annihilation was usually assumed which automatically took care of the charge conservation, since the atom as a whole is neutral and the photons do not carry charge. This idea was soon replaced by an electron-positron pair process when it became known that the primaries are charged. Again charge conservation does not lead to any difficulty. Recently it has been necessary to replace this in favor of a two-proton annihilation since the primaries are protons. But a two-proton annihilation is obviously not possible for a neutral atom because charge would not be conserved unless we assume some kind of highly artificial way out, such as a simultaneous production of two low-energy electrons.

There are other difficulties of an even more serious nature. The maximum energy of protons which could be observed with any reasonable frequency would be the energy of the silicon annihilation process and would amount to 13.2 Bev for each proton. Cosmic ray protons occur with much higher energy than this. Even the heaviest element would give at most an energy of 10^{11} ev for each proton, which is still many powers of ten less than the maximum energy observed in cosmic ray protons. It would thus be necessary to assume another process for a source of the very high energy protons, such as, for instance, the simultaneous annihilation of a large number of atoms.

A further difficulty results from the fact that the annihilation hypothesis leads to a probability for the annihilation which is much too large to have escaped detection. For the special case of silicon, for instance, a decay time of 2×10^{24} sec is obtained on the basis of the following extremely conservative assumptions: No radiation escapes the galaxy and no radiation is absorbed inside the galaxy. The density of silicon was constant in the past and amounts to 10^{-4} atoms/cm^3; only 1% of the cosmic radiation is silicon radiation; (actually the radiation with energy larger than 10 Bev is about 5 to 10% of the total radiation) and finally the time of existence of the galaxy is 10^{17} sec. Such a decay time would no doubt lead to detectable effects in the laboratory. No such effects have ever been found. It is difficult to take seriously the proposal made by Millikan that for some unexpected reason the annihilation should only proceed when matter is in the

extremely rarefied state of interstellar space, while it should not occur under ordinary laboratory conditions.

A still further difficulty is found in the occurrence of heavy ions in the primaries with an abundance which is at least comparable with their natural relative abundance in the universe.

In spite of these difficulties it has been repeatedly claimed by Millikan and co-workers that their measurements of the variation of the intensity with geomagnetic latitude definitely proves the existence of the plateau character of this intensity when plotted as a function of geomagnetic latitude. It is conceivable that a sympathetic interpreter of these measurements might see some kind of irregularity in their measuring points which is not inconsistent with such a character of the curve. However, the errors in the measuring points are so large that the curve may just as well be assumed smooth. Kusaka (*50*) has actually carried out an evaluation of the intensity curve as a function of geomagnetic latitude under the assumption of a power law for the primary spectrum and finds the agreement with the measuring points of Millikan, Neher and Pickering quite satisfactory.

We conclude, therefore, that there is no unambiguous experimental evidence for the atom annihilation hypothesis and that it leads into serious theoretical difficulties which could only be alleviated with a host of further assumptions that have no basis in fact.

Similar difficulties are encountered in a variant of the annihilation hypothesis such as the one discussed by Rojansky (*51*) and Klein (*52*). Here the existence of reversed matter in the universe is postulated composed of atoms with antiprotons and antineutrons in the nucleus and positrons instead of electrons surrounding them. No experimental evidence for such matter exists, and the theoretical difficulties are similar as for the ordinary annihilation process.

We turn now to the theories which see the origin of cosmic rays in some kind of acceleration process. Since a magnetic field alone cannot accelerate a particle, there must be electrical fields present. The potential difference for such an accelerating field must range to values as high as 10^{17} volts. Since no appreciable electric field strength has been observed in the stellar spectra, it follows that the acceleration process must take place over large distances, possibly even in several stages.

An acceleration process of this sort was discussed by Alfvén (*53*). He shows that, if the components of a double star have magnetic moments inclined to the axis of rotation of the system, then an alternating electric field in the direction of the axis will result which may accelerate ions to as much as 10^{14} ev for reasonable assumptions about the strength of the dipole moment and the separation of the components.

The main difficulty with this theory, and in fact with any acceleration theory, is that it is impossible to account for the actual intensity of cosmic rays. This can be seen easily as follows: Let us assume that the radiation is confined to the interior of the galaxy due to the action of a magnetic field. The known intensity of the primaries is 0.12 particles cm^{-2} sec^{-1} $sterad^{-1}$. Each of these particles carries in the average a kinetic energy of about 20 Bev. From this it follows that cosmic rays fill the interstellar space with an average energy density of about 5×10^{-12} erg cm^{-3}. The total energy in the galactic system of volume 10^{68} cm^3 is, therefore, 5×10^{56} erg. This energy is not a permanent reservoir since it is being continually dissipated.

The most important process of dissipation which outweighs all the other effects is the slowing down due to mesotron production in the collision with the dif-

fuse matter in interstellar space. This is the same process which eliminates the primary radiation in the atmosphere. It is known from cosmic-ray measurements that the effective depth of the atmosphere for the elimination of the primaries is about 100 gm cm^{-2}. Thus we would expect that a cosmic-ray particle would lose its energy in interstellar space after traveling a thickness of about 100 gm cm^{-2}. Since the density of interstellar space is about 10^{-24} gm cm^{-3} and the velocity of a cosmic-ray particle is $c = 3 \times 10^{-10}$ cm sec^{-1}, the energy of the cosmic rays is dissipated in a time of approximately 10^8 years. The sources of cosmic rays must, therefore, replenish the total cosmic-ray energy of 5×10^{56} erg in a time of about 10^8 years. Now the total kinetic energy of all the stars is just about of the same order of magnitude as the total cosmic-ray energy, or 10^{56} erg.

Since Alfvén's acceleration process takes its energy from the orbital motion of the double star, it is clear that the orbital energy would not be enough to furnish energy at the rate which is needed for the cosmic rays. The argument just prepared is a very general one and applies to all types of acceleration processes connected with stellar objects. It seems necessary, therefore, to look for an acceleration process where the energy furnished comes from the diffuse matter in interstellar space.

Another possibility of avoiding the above-mentioned difficulty is to assume that cosmic rays exist only in very small local regions so that the total energy contained in the cosmic rays is much less than the 5×10^{56} erg found before. At the present time there is no mechanism known which would restrict cosmic rays to such regions.

This brief sketch on the speculations regarding the origin of the primaries may suffice to illuminate the fact that very little is as yet known about the origin of the primaries.

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The references below and those which appeared in the first half of this paper published last month are only a small selection of the research papers on cosmic rays.

The principle of selection used follows: In each area some of the early pioneer work and then again the latest work of importance is quoted. Intermediate work is summarized in the text but not quoted.

Further references may be found in the review papers and books quoted at the end of this list.

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The author is very much indebted to Professor M. Schein for reading the manuscript and commenting on various points. Thanks are also due to J. T. Curtis for some help in sifting the literature.

BOOKS

INTRODUCTION TO ATOMIC PHYSICS, by Otto Oldenberg, McGraw-Hill Book Co., Inc., New York, 1949, 373 pages, $5.00. *Reviewed by Keith Henney, Consulting Editor,* NUCLEONICS.

While this book is designed as a text for students who have had one year of physics and who are familiar with the elements of chemistry, there is no reason why it should not be very useful for those countless engineers and others whose formal training occurred many years ago. This reviewer read the entire book with pleasure and with a degree of comprehension that depended more upon the reader than upon the writer. Anyone desiring to get an accurate and broad view of nuclear matters without getting in over his head will find Professor Oldenberg's book good reading.

It is divided into seven broad parts which are not only logical but to some extent chronological. Thus the first deals with the structure of matter as revealed in chemistry; then follows sections on gases, on the structure of electricity, the structure of light, the electronic structure of atoms, nuclear structure and the wave nature of matter. At the end of each chapter is given a series of problems which serve not only to check one's knowledge but to teach more of the facts of the matter under discussion.

The reader may find the chapter on "Applications" very interesting since it indicates the usage to which nuclear knowledge has been and will be put.

ISOTOPIC CARBON by M. Calvin, C. Heidelberger, J. C. Reid, B. M. Tolbert, and P. E. Yankwich, published by John Wiley & Sons, Inc., New York, 1949, 376 pages, $5.50. *Reviewed by A. K. Solomon, Harvard Medical School.*

This book is a first-class laboratory manual written by five members of the scientific staff of the University of California's Radiation Laboratory. In reading through the book, one is increasingly aware that it represents the careful conclusions of a group well seasoned in the techniques and prob-

lems involved in experiments with carbon isotopes. They have chosen to write about carbon because it plays so prominent a role both in chemistry and biology; it happens also to possess a remarkably complete array of isotopic tracers, including the stable isotope, carbon-13, and the two radioactive ones, carbons-11 and 14. Thus, while the book describes only techniques involving a single element, it includes many of the known methods of measurement, for carbon-13 demands a mass spectrometer, and carbon-14 demands the rather specialized counting techniques required for the measurement of isotopes with very soft beta radiation.

Since carbon is so important an element, the results of research involving it are scattered in many publications. One of the most useful functions performed by the book is the collation of this varied material in chapters concerned first with the chemical synthesis of tracer-marked carbon compounds, then with their degradation, and finally with biosynthetic means of preparation. A ten-page bibliography summarizes all the published work involving tracer carbon. The easy reference that results from the collection of this material is particularly welcome. The special techniques involved in handling these compounds are well illustrated by a chapter on vacuum techniques in organic chemistry. If the book contained no more than these chemical bones, it would still be a most important compendium.

A technique such as the use of tracers in chemistry and biology attracts many investigators, because it appears at first sight to present an easy means of getting at truths hitherto hidden. There is no doubt that it does present a way of getting at such new truths, but there can also be no doubt to the thoughtful reader of this volume that the way is not easy. The problems in isotope research are often peculiar, and the pitfalls are all too many. Since radioisotopes can be detected in such small quantities, it might seem that the method would make microanalysis particularly simple. However, the reverse is in many cases the fact, for small quantities of isotope can be absorbed or carried down in the most unlikely precipitations. As the authors point out, it is absolutely essential to make sure of the isotopic purity of compounds being identified by their radioactivity. They prove their point by an absolutely hair-raising example of co-precipitation.

For the novice, there are presented enough of the elementary details of counting and measurement that he can feel confident in setting up the necessary apparatus. For the amateur, there are made available the details of many of the more advanced techniques, including counting in the gas phase, and the use of the Lindemann and vibrating-reed electrometers. For the professional there is a set of four appendices in which he can exercise his mathematical faculties. Happily, exercises of this sort have been relegated to the back of the book, and the equations need not frighten the non-mathematical reader. A good deal of its especial value as a laboratory manual springs from the complete presentation of all the considerations required for the measurement of a single element.

"Isotopic Carbon" is a fine handbook, of great value to the scientist who works with any isotope, and invaluable for the man who works with carbon.

NUCLEONIC EVENTS

AEC SELECTS IDAHO SITE FOR REACTOR TEST STATION

A 400,000 acre site near Pocatello, Idaho, has been chosen by the Atomic Energy Commission for its new reactor testing station. In area, the tract is about the same as that of the Hanford plutonium production plant at Richland, Wash.

Construction of the new test station and its nuclear reactors will cost about $500 million, according to Congressional estimate. It is expected that 6,000 workers will be employed at the peak in building the facilities. The station will serve as a testing and proving plant for the commission's entire reactor development program.

For the present, it has been definitely decided to locate at the station two of the four reactors that are now in the design stage. One of the two is a materials testing reactor, and the other is the Navy's reactor for ship propulsion. Additional reactors will be constructed at the station in the future.

Of the land selected for the site, all but about 20,000 acres belongs to the government. The federal property includes the 173,000-acre Navy Proving Ground at Arco—not in active use at present. Negotiations are in progress for transfer of the Navy property to the AEC. Engineering surveys of the entire test area are nearly complete, but detailed boundaries have not yet been fixed.

Lester E. Johnston, manager of the Knolls Atomic Power Laboratory in Schenectady, has been appointed manager of the test station. He has been associated with the atomic energy program since 1944. At that time he was in the Army Corps of Engineers and assigned to Oak Ridge. He was construction officer for the electromagnetic plant which produced uranium-235 during the war. Later he was an assistant unit chief at the Oak Ridge gaseous diffusion plant, now the chief source of uranium-235 for the national atomic energy program. In June, 1946, Mr. Johnston opened the Schenectady area office when the Knolls Atomic Power Laboratory project was started with the General Electric Company as the operating contractor.

CANADIAN SCHOOL OFFERS NUCLEAR CHEMISTRY COURSE

The chemistry department of Dalhousie University, Halifax, Nova Scotia, with the cooperation of the Nova Scotia Research Foundation and the National Research Council of Canada, is offering a six weeks summer course (July 4 to August 13) in applied nuclear chemistry and tracer techniques. This course is announced as being equivalent to one regular university graduate course and is designed primarily for graduate chemists and chemical engineers. A necessary prerequisite is a working knowledge of general analytical chemistry. Daily lectures are to be supplemented by seminars and problem periods. Laboratory work includes studies of different methods and instruments for assaying, utilizing and safely handling a wide variety of radioactive isotopes. Experimental studies of various applications of radioactive tracers are based largely on published researches and declassified government documents.

Only a limited number of applicants

can be accommodated. Inquiries for further information should be addressed to the Registrar, Dalhousie University, Halifax, N. S.

NRC PUBLISHES FOURTH PRELIMINARY REPORT

Preliminary report No. 4, "Monoenergetic Neutrons from Charged Particle Reactions," by A. O. Hanson and R. F. Taschek, was published last month by the Committee on Nuclear Science of the National Research Council. Issued previously, and designated as preliminary reports 1, 2 and 3, respectively, were "Isotopic Weights of the Fundamental Isotopes," by K. T. Bainbridge; "Nuclear Electric Quadrupole Moments and Quadrupole Couplings in Molecules," by B. T. Feld; and "Neutrons from Alpha Emitters," by H. L. Anderson.

Copies of these reports may be obtained from Dr. R. C. Gibbs, Division of Physical Sciences, National Research Council, 2101 Constitution Avenue, Washington 25, D. C.

A fifth preliminary report, "Energy Levels of Light Nuclei," by T. Lauritsen, is now being prepared by the Committee for publication.

AEC ESTABLISHES PATENT COMPENSATION BOARD

The Atomic Energy Commission recently established a Patent Compensation Board to consider and act on applications arising under the patent and invention provisions of the Atomic Energy Act. So far as these provisions prevent the issuance of patents in accordance with the regular patent system, the AEC is empowered to make awards after hearings held before the new board.

Members of the board are: Casper W. Ooms, who has served as a member of the AEC's Patent Advisory Panel since January, 1947; John V. L. Hogan, a consulting engineer, and president of the Interstate Broadcasting Company and Radio Inventions, Inc.; and Isaac Harter, chairman of the Babcock and Wilcox Tube Company and a member of the recent AEC Industrial Advisory Group.

LATTES MAY RECEIVE $25,000 PRIZE FOR MESON WORK

Brazilian nuclear physicist Cesar Lattes would be given a $25,000 prize for his work in production of artificial mesons at the University of California, under a bill which is expected to pass the Brazilian Congress. The same bill would grant $150,000 to the University of São Paulo, where Lattes is to work, to equip the physics department of the Faculty of Philosophy, Science and Letters for continued investigations in nuclear physics.

"BUSINESS WEEK" REPORTS ON ATOMIC ENERGY PROGRAM

Business Week's third annual report on the nation's atomic energy program appeared in the April 30 issue of the weekly magazine. As in the past, Robert Colborn has reported on the results of his visits to all of the major AEC laboratories and his discussions with top-level officials in the AEC and industry. Single copies of this report are available at 20 cents apiece. Orders should be addressed to Readers Service Dept., *Business Week*, 330 W. 42nd St., New York 18.

SCINTILLATION COUNTER SYMPOSIUM PLANNED

A symposium on scintillation counters will be held in Oak Ridge on June 3 and 4. Invited papers will be presented by H. Kallman, F. Seitz, G. A. Morton and P. R. Bell. Martin Deutsch, Massachusetts Institute of Technology, will serve as chairman. All who are interested are invited to

REPRINTS OF "INSTRUMENT HANDBOOK" AVAILABLE

A limited number of extra copies of "Nuclear Instrument Handbook," the special section appearing in this issue, are available at 50 cents apiece. Cash should accompany orders, which should be addressed to The Editor, NUCLEONICS, 330 West 42nd St., New York 18.

attend. Further details may be obtained from W. H. Jordan, Oak Ridge National Laboratory, Oak Ridge, Tenn.

NOTES ON PROSPECTING

Britain is making special efforts to encourage mining houses and prospectors in its colonies to locate deposits of the radioactive materials, uranium and thorium. It is considered that there are more chances of important deposits being found there than in Britain, which, it is said, has already been surveyed more completely than any other part of the world.

The British Ministry of Supply is publishing in all likely territories an offer to buy "all uranium ores and concentrates produced in the Colonial Empire during the next ten years, at a minimum price of 13/9d. ($2.75) a pound of contained uranium oxide delivered." This applies to lots of at least ten tons, containing at least ten percent uranium oxide.

Higher prices may be fixed by negotiation where costs of production are greater or where there are other valuable constituents in the ore. The Ministry also may be willing to provide capital to install concentrating plants and, in some cases, pay a lump sum in aid of mine development. It will discuss terms for the purchase of thorium-containing materials. Similar prices would be paid for any uranium found in Britain.

Colonial geological surveys and the Geological Survey of Great Britain are giving advice and analyzing samples brought in by prospectors, and instruments for measuring radioactivity are being sold in increasing numbers in the colonies. The search for radioactive minerals in the Dominions is already being actively pursued.

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Argentina has announced the discovery of a new uranium vein which promises to be the most important yet found in the country.

Located in the Andean foothills of the northwestern province of Catamarca, government officials say the vein is just over 50 yards wide and presents outcroppings of uranium ore over an area of roughly 25 square yards. The strike is about 30 miles from a previously discovered pitchblende deposit.

The vein was unearthed by a Swedish engineer, Gunnar Jarnheim, and his associate, Alberto Lapidus, in the course of mica mining operations. The uranium content of samples proved high in initial studies by the provincial school of mines, while Argentine authorities announced that "the first analyses permit the expectation of a combination different from any classified at present."

Under the terms of an Argentine law passed late in 1946, the discoverers are entitled to a bounty of $4,000 to $60,000, depending on the yield of the vein. The same law nationalized all radioactive materials in Argentina.

The South American country is already working small uranium deposits in the provinces of Córdoba, San Luis and Mendoza.

NUCLEAR NEWSMAKERS

Philip M. Morse, professor of physics at Massachusetts Institute of Technology and former director of Brookhaven National Laboratory, has been appointed research director and deputy

director of the Weapons Systems Evaluation Group of the National Military Establishment. Recently created, this group is responsible for the technical and operational evaluation of all present or projected systems of attack and defense.

Rear Admiral **T. B. Hill,** an expert on gunnery and fire control, has been designated director of atomic energy for the Navy to replace Rear Admiral **W. S. Parsons,** who is now with the Weapons Systems Evaluation Group.

Paul Wang, former dean of the physics department at Shantung University, has joined the staff of the X-ray laboratory of the National Bureau of Standards, where he will be concerned with research in X-rays and nuclear physics, particularly betatron and high-voltage equipment. His present investigations include the design and development of a new type of ionization measurement equipment, X-ray dosage research, and experimental verification of modern theories of high-energy radiation.

A committee on equipment and control to advise the export control branch of the Atomic Energy Commission has been established. The members of the committee include: **R. W. Albright,** Distillation Products, Inc., Rochester, N. Y.; **A. O. Beckman,** National Technical Laboratories, S. Pasadena, Calif.; **G. H. Bucher,** Westinghouse Electric Corp., Pittsburgh, Pa.; **C. S. Redding,** Leeds & Northrup, Philadelphia, Pa.; **J. A. Victoreen,** Victoreen Instrument Co., Cleveland, O.; **R. S. Morse,** National Research Corp., Cambridge, Mass.; and **H. B. Neal,** Kinney Mfg. Co., Boston, Mass.

The Atomic Energy Commission has established a permanent personnel security review board to succeed the original board which completed its work last year. Members of the new board are: **Charles Fahy,** an attorney, of Washington, D. C., who will serve as chairman of the board; **Arthur S. Flemming,** president of Ohio Wesleyan University; and **Bruce D. Smith,** director of the United Corp., New York, N. Y., and the Lehigh Coal and Navigation Co., Philadelphia, Pa.

The former Committee on Standards of Radioactivity, later known as the Committee on Radioactivity, of the Division of Physical Sciences of the National Research Council has been reconstituted as a Committee on Nuclear Science. Its responsibilities have been enlarged to include work on units, standards, and measuring instruments in this field. The membership of the new committee includes: **L. F. Curtiss,** National Bureau of Standards, chairman; **R. D. Evans,** Massachusetts Institute of Technology, vice chairman; **Irving Feister,** National Bureau of Standards, secretary. The following are chairmen of the subcommittees indicated: **H. L. Anderson,** Neutron Measurements and Standards; **L. F. Curtiss,** Beta- and Gamma-Ray Measurements, and Units; **R. D. Evans,** Shipment of Radioactive Substances; **H. H. Goldsmith,** Publications and Information; **J. G. Hamilton,** Radiobiology; **P. M. Hurley,** Radiogeology; **J. B. H. Kuper,** Instruments and Techniques; **W. M. Manning,** Radiochemistry; and **W. H. Sullivan,** Nuclear Constants. The liaison members of the committee are **P. C. Aebersold** for the Atomic Energy Commission; **T. J. Killian** for the Office of Naval Research; **Herbert Scoville, Jr.,** for the Armed Forces Special Weapons Project; and **W. D. Urry** for the Air Force.

Cleveland Norcross, general manager of the American Institute of Physics, died on March 21 after an emergency operation for a brain tumor. Mr. Norcross had been in his position with the Institute since July, 1947.

ABSTRACTS

CHEMICAL PUBLICATIONS

The kinetics of the oxidation of 2-deuteropropanol-2 by chromic acid, F. H. Westheimer, N. Nicolaides (Univ. of Chicago, Ill.), *J. Am. Chem. Soc.* **71,** 25–28 (1949). The deuteration of acetone was found to be accompanied by some hydrogen-deuterium exchange, so that the deuteration product was a mixture containing 55% of 2-deuteropropanol-2 (*I*). Oxidation of *I* by chromic acid was only about one-sixth as fast as is the oxidation of ordinary isopropyl alcohol. It follows that the secondary hydrogen atom is removed during the rate-controlling step of the reaction.

A method for the determination of organic compounds in the form of isotopic derivatives—I. Estimation of amino acids by the carrier technique, A. S. Keston, S. Udenfriend, R. K. Cannan (New York Univ., N. Y.), *J. Am. Chem. Soc.* **71,** 249–257 (1949). The components of a mixture of related organic compounds may be estimated by an isotope dilution method. The mixture is treated with an isotopic reagent, which converts the components quantitatively to isotopic derivatives. After the addition of a large excess of the unlabeled derivative of the desired constituent, as a carrier, to the mixture, the derivative is separated, purified, and counted. The method was applied to determine the glycine, alanine, and proline in a mixture of twelve synthetic amino acids, and in the crystalline proteins β-lactoglobulin, human hemoglobin, aldolase, and phosphoglyceraldehyde dehydrogenase. The isotopic reagent used was *p*-iodophenyl sulfonyl chloride-I^{131}.

Thermal exchange experiments with radioactive zinc. L. Levanthal, C. S. Garner (Univ. of California, Los Angeles), *J. Am. Chem. Soc.* **71,** 371 (1949). A study was made of the exchange of 250-day Zn^{65} between dipyridine zinc acetate and the following zinc compounds in pyridine solution at 25°: zinc acetylacetone, zinc acetylacetone ethylenediimine, zinc benzoylacetone ammoniate, zinc nicotinylacetone, and dipyridine zinc thiocyanate. Complete exchange was found to occur in each case after exchange times of about 30 seconds. If rapid exchange was not induced by the separation process used, the zinc complex compounds studied were not considered suitable for concentrating Zn^{65} by the Szilard-Chalmers method. These compounds are apparently unstable with respect to ionization or displacement reactions.

Thermal exchange experiments with radioactive chromium, H. E. Menker, C. S. Garner (Univ. of California, Los Angeles), *J. Am. Chem. Soc.* **71,** 371–372 (1949). The exchange reaction between chromium (III) and dichromate ions in sulfuric acid, perchloric acid, and sodium hydroxide solutions was studied using 27-day Cr^{51}. Also investigated was the exchange of chromium (III) with hexathiocyanatochromate (III), hexacyanochromate (III), and trioxalatochromate (III), respectively, in various solvents. Appreciable exchange was observed only for $Cr(III)-Cr(CN)_6^{=}$ in low concentration of perchloric acid or acetic acid, and for hexaaquochromium (III) and dichromate ions in 0.05*f* perchloric acid at 25° and 45°.

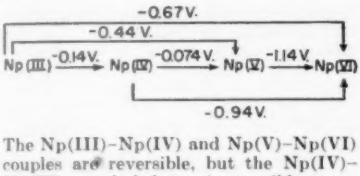
Preparation of radioactive CO_2 from $BaCO_3$, N. Zwiebel, J. Turkevich, W. W. Miller (Brookhaven Natl. Lab., Upton N. Y.), *J. Am. Chem. Soc.* **71,** 376–377 (1949). C^*O_2 may be obtained from BaC^*O_3 by using a thermal reaction between dry solids, such as the reaction between BaC^*O_3 and $PbCl_2$. The lead carbonate is unstable and decomposes at a low temperature. At 400° the reaction is 96% complete in four hours.

The use of radioactive tracers in a study of activator distribution in infrared-sensitive phosphors, R. W. Mason, C. F. Hiskey, R. Ward (Polytechnic Inst. of Brooklyn, N. Y.), *J. Am. Chem. Soc.* **71,** 509–514 (1949). Radioactive isotopes of europium, cerium, and samarium were used to study the distribution of these rare earth ions between the phosphor base materials, strontium sulfide and strontium selenide, and saturated solutions of these bases in the fluxes, strontium chloride and lithium fluoride. The intimately mixed phases were separated by a procedure which involved filtration at the fluxing temperature (about 1000°) using a platinum Munroe crucible.

A synthesis of adenine. The incorporation of isotopes of nitrogen and carbon, L. F. Cavalieri, J. F. Tinker, A. Bendich (Sloan-Kettering Inst. for Cancer Research, New York), *J. Am. Chem. Soc.* **71,**

533–536 (1949). Earlier procedures for the synthesis of adenine labeled with N^{15} in the 1 and 3 positions were reinvestigated, and an improved synthesis was evolved. The introduction of C^{13} or C^{14} into the 4 and 6 carbons of the adenine, as well as into malononitrile and phenylazomalononitrile was accomplished.

The oxidation states of neptunium in aqueous solution, J. C. Hindman, L. B. Magnusson, T. J. LaChapelle (Argonne Natl. Lab., Chicago, Ill.), *J. Am. Chem. Soc.* **71,** 687 (1949). The existence of +4, +5, and +6 states of neptunium was shown by means of spectrophotometric titrations of neptunium solutions with oxidizing and reducing agents. Isolation and X-ray identification of the salt $NaNpO_2(OOCCH_3)_2$ from the solution of the highest oxidation state served to establish the identity of one oxidation state. The +5 state is quite stable in acid solution. The formal oxidation potentials (not E^0 values) were determined in one molar hydrochloric acid and may be represented by the following scheme:



The Np(III)–Np(IV) and Np(V)–Np(VI) couples are reversible, but the Np(IV)–Np(VI) couple behaves irreversibly.

A method of estimating and minimizing the error of measurement of the rate of a radioactive exchange reaction, N. Davidson, J. H. Sullivan (California Inst. of Technology, Pasadena) *J. Am. Chem. Soc.* **71,** 739–740 (1949). In a radioactive exchange reaction between two components in chemical equilibrium, the change in activity of these components with time introduces an error in the measured rate constant. A method is described of estimating this error in the rate constant as a function of the extent of exchange. By selection of the optimum degree of exchange the error may be minimized.

Comparison of age with relative abundance of argon and potassium in rocks, R. L. Farrar, Jr., G. H. Cady (Univ. of Washington, Seattle) *J. Am. Chem. Soc.* **71,** 742–743 (1949). A number of rocks of various known geologic age were analyzed for helium, argon, and potassium. The results indicate no regular increase of the A/K ratio with age. The range of argon content was much less than that of helium. Without an isotopic analysis to establish the relative proportions of atmospheric and K^{40}-derived argon, the ratio A/K is not a good measure of the age of a rock.

. . . I. W. RUDERMAN

LIFE SCIENCE PUBLICATIONS

Interaction of calcium, phosphorus and vitamin D—III. Study of mode of action of vitamin D using Ca^{45}, B. Migicovsky, A. Emslie (Vitamin and Physiol. Research Lab., Experimental Farm, Ottawa, Can.), *Arch. Biochem.* **20,** 325–330 (1949). Experiments indicate that vitamin D decreases the loss of calcium via excretion in starving chicks but has little effect on phosphorus. Calcium ingestion decreases the loss of phosphate. Vitamin D exerts a retarding effect on the loss of calcium from newly formed bone.

The use of radioactive isotopes in immunological investigations—II. The fate of injected P^{32}-containing proteins, T. Banks, J. Boursnell, H. Dewey, G. Francis, R. Tupper, A. Wormall (Dept. of Biochem., Med. Coll. of St. Bartholomews Hosp., London), *Biochem. J.* **43,** 518–523 (1949). Injected P^{32}-labeled lipovitellin, vitellin and artificially phosphorylated human blood serum disappear rapidly from the blood stream. Three quarters of the phosphorylated human blood serum disappears three hours after injection into rabbits.

Radiocardography and its clinical applications, M. Prinzmetal, E. Corday, R. Spritzler, W. Fleig (Inst. for Med. Research, Cedars of Lebanon Hosp., Los Angeles, Calif.), *J. Am. Med. Assoc.* **139,** 617–622 (1949). Radiocardiography using Na^{24} has been used to determine the pumping qualities of the heart.

Rate of elimination of radioactive carbon administered as carbonate from the tissues and tissue components of mature and growing rats, J. Schubert, W. Armstrong (Dept. of Physiol. Chem., Univ. of Minnesota, Minneapolis), *J. Biol. Chem.* **177,** 521–527 (1949). The specific activity of C^{14} in tissues and tissue components of growing rats greatly exceed in general those of mature rats. Practically no change in over-all C^{14} retention occurred after the fifteenth day in mature rats, whereas a significant over-all decrease in C^{14} retention was observed in the growing

animals. After the eighth day the biologic half-life of the C^{14} in intact and inorganic bone was 12 to 15 days in growing animals. In mature rats it was 30 to 40 days. The specific activity of serum proteins and the rates of decrease of C^{14} content were similar for both groups. The rate of decrease of C^{14} in the soft tissues of mature rats fell appreciably by the twentieth day. No significant C^{14} turnover in the brain protein of young rats was noted.

A synthesis of isotopic cytosine and a study of its metabolism in the rat, A. Bendich, H. Getler, G. Brown (Sloan-Kettering Inst. for Cancer Research, New York), *J. Biol. Chem.* **177,** 565–570 (1949). Cytosine has been synthesized containing isotopic nitrogen in the 1 and 3 positions from urea and cyanoacetaldehyde diethyl acetal in over-all yield of 56%. It was shown that cytosine is apparently not a specific metabolic precursor of nucleic acid nitrogen.

Biosynthesis of uric acid labeled with radioactive carbon, J. Karlsson, H. Barker (Division of Plant Nutrition, Univ. of California, Berkeley), *J. Biol. Chem.* **177,** 597–599 (1949). Uric acid prepared biologically by the use of pigeons has been labeled in the 2 and 8, the 4, and the 6 positions starting with labeled formate, carboxyl-labeled glycine and labeled bicarbonate, respectively. Uric acid could not be labeled solely in the 5 position from methylene-labeled glycine because some isotope was found in the 2, 4, 6, and 8 positions when this was attempted.

The distribution of fixed radioactive carbon in glucose from rat liver glycogen, W. Schreeve, G. Feil, V. Lorber, H. Wood (Dept. of Biochem., Western Reserve Univ., Cleveland, O.) *J. Biol. Chem.* **177,** 679–682 (1949). Glucose from rat liver glycogen after administration of labeled bicarbonate contained isotope in the 3 and 4 carbons. Trace amounts were found in the other carbons and possible explanations for their presence were advanced.

Incorporation of isotopic carbon dioxide in rat liver glycogen *in vitro,* A. Hastings A. Solomon, C. Anfinsen, R. Gould, I. Rosenberg (Dept. of Biol. Chem., Harvard Med. School, Boston, Mass.), *J. Biol. Chem.* **177,** 717–726 (1949). Carbon dioxide incorporation averaged about 8% in liver glycogen of fasted animals. It was less in well-fed animals or when glucose was substituted for pyruvate as a substrate. Potassium ions were found to favor glycogenesis while sodium ions favored glycogenolysis. Glycogen formation and carbon dioxide incorporation were greater during incubation when both sodium and potassium ions were present in equimolar amounts than when either one was present alone.

Metabolism of isotopic pyruvate and acetate in rabbit liver slices *in vitro,* R. Gould, A. Hastings, C. Anfinsen, I. Rosenberg, A. Solomon, Y. Topper (Dept. of Biol. Chem., Harvard Med. School, Boston, Mass.), *J. Biol. Chem.* **177,** 727–731 (1949). C^{14}-carboxyl-labeled acetate and C^{14}-α-carbon-labeled pyruvate were prepared. A comparison was made of labeled carbon incorporated into rabbit liver glycogen *in vitro* in the presence of the carboxyl-labeled acetate, α-carbon-labeled pyruvate and carbon dioxide. Pyruvate, it is believed, will more readily equilibrate with carbon dioxide than convert itself to glycogen in livers of unfasted rabbits. It is believed that acetate may go to glycogen *in vitro* without prior combustion to carbon dioxide.

A study of transmethylation with methionine containing deuterium and C^{14} in the methyl group, E. Keller, J. Rachelle, V. du Vigneaud (Dept. of Biochem., Cornell Univ. Med. Coll., New York), *J. Biol. Chem.* **177,** 733–738 (1949). Deuterium and C^{14}-labeled L-methionine was fed to a rat and choline and creatine isolated from the body. The ratio of deuterium to C^{14} in the methyl groups of creatine and choline was the same as in the dietary methionine.

Oxidation-coupled incorporation of inorganic radiophosphate into phospholipide and nucleic acid in a cell free system, M. Friedkin, A. Lehninger (Dept. of Biochem., Univ. of Chicago, Ill.), *J. Biol. Chem.* **177,** 775–788 (1949). The oxidation of *l*-malate via the Krebs tricarboxylic acid cycle in the presence of inorganic P^{32}-labeled phosphate, adenine nucleotide, and Mg^{++} in a suspension of particulate material of rat liver, largely of nuclei and mitochondria in make-up, resulted in P^{32}-incorporation into phospholipide, pentose nuclei acid, and an unidentified phosphoprotein fraction of the particulate material. The rate of incorporation of radioactive phosphorus into acid-insoluble esters was much lower than into the acid-soluble fraction.

Carbamates in the chemotherapy of leucemia—IV. The distribution of radioactivity in tissues of mice following injec-

tion of carbonyl-labeled urethane, C. Bryan, H. Skipper, L. White, Jr. (Southern Research Inst., Birmingham, Ala.), *J. Biol. Chem.* **177,** 941–950 (1949). Within 24 hours following intraperitoneal injection, 90% of the labeled carbonyl carbon was expired as carbon dioxide; 5% to 10% was found in the urine. Of the remaining radioactive carbon the highest concentrations were found in the liver, whole blood, intestines and tumor tissue. Two mice with advanced spontaneous lymphoid leucemia and two with mammary carcinoma retained much more radioactive carbon in all tissues after 24 hours than did the normal mice. Whether this was due to failure to catabolize the urethane as rapidly as normal mice, or whether it was due to a greater fixation of urethane or one of its metabolites, was not determined.

The metabolism of carrier-free radioberyllium in the rat, J. Crowley, J. Hamilton, K. Scott (Crocker Lab., Univ. of California, Berkeley), *J. Biol. Chem.* **177,** 975–984 (1949). Within 24 hours, 40% of the injected radioberyllium was absorbed and 54% of this amount excreted mainly in the urine. After 64 days, 80% of the dose had been absorbed and of it 70% had been excreted. Of the remaining Be^7 the bulk was taken up by bone which maintained a level of about 27% during the 64-day period. Liver and kidney Be^7 levels initially compared with bone but fell off more than tenfold after 64 days. Less than 0.2% of an orally administered dose of Be^7 was absorbed by the intestinal tract.

The *in vitro* incorporation of the methylene carbon of glycine into rabbit bone marrow fats, K. Altman (Dept. of Radiation Biol., Univ. of Rochester, N. Y.), *J. Biol. Chem.* **177,** 985–986 (1949). The methylene carbon of glycine is incorporated into the fatty acids of bone marrow and the isotope concentration of saturated fatty acids exceeds that of unsaturated fatty acids. The experiment suggests that rabbit bone marrow may be capable of effecting the conversion of glycine to acetate which is a known precursor of fatty acids.

The phosphorus turnover of carcinoma of the human stomach as measured with radiophosphorus, J. Schulman, Jr., M. Falkenheim, S. Gray (Biophysical Lab., Harvard Med. School, Boston, Mass.), *J. Clin. Invest.* **28,** 66–72 (1949). The rate of phosphorus turnover by gastric carcinoma is at least 45% greater than by normal gastric mucosa. There was at least a 124% increase in turnover of protein phosphorus and at least 45% in lipid phosphorus; the acid-soluble phosphorus turnover remained constant. The increased turnover rate was limited to cancerous tissue and not to the surrounding tissue. In the normal stomach, there are no differences in those areas in which cancer is common and in which cancers are rare. No difference between the phosphorus content of the normal and cancerous mucosa of the stomach was noted.

The distribution in rabbit tissue of intravenously injected iodine as shown by the radioisotope I^{130}, W. Mann, W. Bale, H. Hodge, S. Warren (Dept. of Radiol., Univ. of Rochester, Ill.), *J. Pharmacol. Exptl. Therap.* **95,** 12–17 (1949). The thyroid gland contained high concentrations of iodine while other tissues contained relatively low concentrations. Data suggest that to produce maximal concentrations of I^{130} in the thyroid the total dose of I^{127} should be kept as low as possible.

The effect of anesthetics on the uptake of radioactive phosphorus by human erythrocytes, V. Pertzoff, C. Gemmil (Dept. of Pharmacol., Univ. of Virginia, Charlottesville), *J. Pharmacol. Exptl. Therap.* **95,** 106–115 (1949). The rate of radiophosphorus uptake by human erythrocytes is variable and is apparently not dependent upon food intake. Sodium barbital and ether have a retarding effect on the uptake of phosphate ion by red blood cells. Methadon (in very dilute solution) and urethane are apparently without effect.

Tissue suspensions for estimations of radioactivity, F. Howarth (Dept. of Pharmacol., Univ. of Manchester, England), *Nature* **163,** 249 (1949). Satisfactory suspensions of brominated organic compounds have been obtained by boiling tissue under reflux with lithium hydroxide in 20% ethanol.

The path of carbon in photosynthesis—IV. The identity and sequence of the intermediates in sucrose synthesis, M. Calvin, A. Benson (Radiation Lab. and Dept. of Chem., Univ. of California, Berkeley), *Science* **109,** 140–142 (1949). Radiogram examination revealed that, in photosynthetic experiments of 30 to 90 seconds' duration, the bulk of the incorporated carbon dioxide is found in the phosphoglyceric acids, triose phosphates and the hexose phosphates. The first free carbohydrate to appear is sucrose. It is

believed that two hexose phosphates are the precursors to sucrose. Glucose-l-phosphate is believed to be one of the precursors due to the large amounts of radioactivity found in this compound.

Thermal separation of radiomercury from radiosodium, P. Reaser, G. Burch, S. Threefoot, C. Ray (Dept. of Medicine, Tulane Univ. School of Med., New Orleans), *Science* **109,** 198 (1949). Heating a mixture of isotopic mercuhydrin, a mercurial diuretic, and isotopic sodium chloride in an oven at 250°C for 1 hour and 20 minutes resulted in more than 99% of the mercury's being driven off while the sodium preparation was not appreciably affected.

1-C^{14}-D-glucose and 1-C^{14}-D-mannose, J. Sowden (Radiochem. Lab., Washington Univ., St. Louis, Mo.), *Science* **109,** 229 (1949). 1-C^{14}-D-glucose and 1-C^{14}-D-mannose were synthesized from C^{14}-methanol in about 60% to 70% yield.

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PHYSICAL PUBLICATIONS

A portable radiation monitor, D. G. Wyatt (Univ. of Oxford, England), *J. Sci. Instr.* (London) **26,** 13–17 (1949). The design and construction of a portable radiation monitor of the ionization-chamber d-c amplifier type are given. For X- and gamma-radiation in the range 100 kev to 3 Mev, the most sensitive range gives a 10% meter reading for a 0.01 r/hr intensity. Alpha and beta-ray intensities can also be investigated by using sliding shutters.

Experiments on *n-p* scattering with 90- and 40-Mev neutrons, J. Hadley, E. Kelly, C. Leith, E. Segrè, C. Wiegand, H. York (Univ. of California, Berkeley), *Phys. Rev.* **75,** 351–363 (1949). The angular dependence of the *n-p* cross section was measured for 40- and 90-Mev neutrons by detection of recoil protons from polythene scatterers. The total cross sections at these energies were measured by attenuation methods. The angular dependence is well explained by a central force of the meson type, containing an exchange term, and corresponding to a meson of mass 326 electron masses.

Stars in photographic emulsions initiated by deuterons—I. Experimental, E. Gardner, V. Peterson (Univ. of California, Berkeley), *Phys. Rev.* **75,** 364–369 (1949). About 300 stars produced in a desensitized emulsion by the deuteron beam of the 184-in. cyclotron were investigated for the energies of 35 Mev, 90 Mev, 130 Mev, and 190 Mev. The plate registered protons of energy less than about 10–15 Mev, and all heavier charged particles. The average number of prongs for each energy was close to 3.0. There were about three times as many prongs in the forward as in the backward direction of the beam.

Stars in photographic emulsions initiated by deuterons—II. Theoretical, W. Horning, L. Baumhoff (Univ. of California, Berkeley), *Phys. Rev.* **75,** 370–378 (1949). A theory of stars was developed on the basis of a transparency given by the Serber theory and used together with evaporation of nucleons obtained from a statistical model of the nucleus. Probability distribution was computed for the number of prongs per star, assuming approximate values of interactions, binding energies and entropies. The results agree qualitatively with experiment.

Positive mesons produced by the 184-inch Berkeley cyclotron, J. Burfening, E. Gardner, C. M. G. Lattes (Univ. of California, Berkeley), *Phys. Rev.* **75,** 382–387 (1949). A 380-Mev circulating alpha-particle beam incident on a carbon target produced positive and negative mesons. Photographic plates were placed to receive those which come off at a small downward angle (2–5 Mev energy). The relative numbers were about one positive to four negative heavy mesons. The heavy positive mesons are observed to decay into secondary mesons.

On the scattering of protons in hydrogen, C. L. Critchfield, D. C. Dodder (Univ. of Minnesota, Minneapolis), *Phys. Rev.* **75,** 419–425 (1949). An analysis is made of recent data on the scattering of protons by gaseous hydrogen. The *S*-wave scattering is found to agree with that given by an attractive 10.5-Mev square well of radius e^2/mc^2, or with that from a Yukawa potential with a meson mass of 300 electron masses. Combination with repulsive *p*-wave scattering explains the small-angle scattering, but the *p*-wave shifts are several times larger than those from a repulsive potential square well with the same dimensions as the *S* well.

An investigation of brehmsstrahlung by means of nuclear isomerism of indium, W. C. Miller, B. Waldman (Univ. of Notre Dame, Ind.), *Phys. Rev.* **75,** 425–432 (1949). Using the X-rays produced by electrons incident on a gold target, In^{115} targets were raised to metastable states.

The results for thick and thin targets, obtained by measuring the activity produced as the electron bombarding energy is varied, showed that the intensity of an isochromat of X-radiation is constant over electron energies of several hundred kev. The absorption by the indium is shown to be a line absorption. Several activation levels were found, and were detected by using the electrons from the metastable level to which the activation levels decay.

88-Mev gamma-ray cross sections, J. L. Lawson (General Electric Co., Schenectady, N. Y.), *Phys. Rev.* **75,** 433–444 (1949). A portion of the brehmsstrahlung produced by the stopping of electrons from a betatron is selected by an arrangement which gives the energy of the pairs produced by such radiation. The portion near the upper limit of the brehmsstrahlung distribution is used to minimize the effect of energy degradation by scattering in the sample whose cross section is measured by transmission. The results show small discrepancies, roughly proportional to Z^2, from the cross sections predicted by theory. Pair-formation cross-section ratios were obtained; Compton-scattering cross sections calculated from them agree with the Klein-Nishina formula within experimental accuracy.

The angular and lateral spread of cosmic-ray showers, J. Roberg, L. W. Nordheim (Duke Univ., Durham, N. C.), *Phys. Rev.* **75,** 444–457 (1949). The spread of electrons and photons as functions of their energy are given analytically down to the critical energy, and calculated numerically down to 4 Mev in air. The radiative and ionization losses were considered simultaneously.

The application of a magnetic lens spectrometer to the measurement of gamma-radiation from Zn^{65} and Co^{60}, E. N. Jensen, L. J. Laslett, W. W. Pratt (Iowa State Coll., Ames, Ia.), *Phys. Rev.* **75,** 458–465 (1949). The effect of the earth's magnetic field and of the thickness of the radiators used to obtain photoelectrons from gamma rays in a thin lens spectrometer is described. Calibration by means of annihilation radiation and the *F* line of ThB was used.

The recording of electron tracks in photographic emulsions, R. H. Herz (Kodak Ltd., Wealdstone, Harrow, Eng.), *Phys. Rev.* **75,** 478–485 (1949). Electron tracks were produced in photographic emulsions by X-rays, natural radioactive decay, weak induced activity, and exposure in an electron microscope. A tentative range-energy relation was investigated by length measurement of the tracks produced by monoenergetic electrons in the microscope and by X-rays (heterogeneous and monochromatic).

Beta-decay of Be^{10} and theory of beta-decay, R. E. Marshak (Univ. of Rochester, N. Y.), *Phys. Rev.* **75,** 513–515 (1949). If the $Be^{10} \rightarrow B^{10}$ decay involves a spin change of three, the minimum half-life observed indicates that the interactions possible on this basis should all give beta spectra very different from the allowed shape.

The beta-spectrum of Be^{10}, D. J. Hughes, C. Eggler, C. M. Huddleston (Argonne Natl. Lab., Chicago, Ill.), *Phys. Rev.* **75,** 515–518 (1949). Using a method by which the absorption curve for Be^{10} is compared with the absorption curves of emitters whose spectra are well known, the experimental Be^{10} spectrum is obtained. The result agrees much better with the allowed shape than with the shape calculated by Marshak on the basis of the half-life and spin change.

Relative cross sections for nuclear reactions induced by high-energy neutrons in light elements, W. J. Knox (Univ. of California, Berkeley), *Phys. Rev.* **75,** 537–541 (1949). Light elements from carbon to sulfur were bombarded with 90-Mev neutrons, and decay curves were taken. Identification of the components of the decay allows a comparison of the yields, and therefore of the cross sections, relative to the yield of a carbon monitor bombarded in the same beam. Cross sections for reactions differing by the emission of an alpha particle or its equivalent are different by a factor of about three.

Magnetic refraction of neutrons at domain boundaries, D. J. Hughes, M. T. Burgy, R. B. Heller, J. W. Wallace (Argonne Natl. Lab., Chicago, Ill.), *Phys. Rev.* **75,** 565–569 (1949). Small-angle scattering of neutrons in unmagnetized iron was investigated. The variation with angle and iron thickness agrees with the theory based on magnetic scattering of the neutrons at domain boundaries.

Thresholds for several photo-nuclear reactions, J. McElhinney, A. O. Hanson, R. A. Becker, R. B. Duffield, B. C. Diven (Univ. of Illinois, Champaign), *Phys. Rev.* **75,** 542–554 (1949). Excitation curves were taken by various methods (varying

for different elements) for bombardment with X-rays from betatron electrons. Calibration was by means of the deuterium, beryllium, carbon, and nitrogen thresholds. The thresholds of various (γ,p) and (γ,n) reactions were determined, and the excitation curves (up to 22 Mev) appear similar in shape, with the exception of that for Ta. The thresholds are correlated with the packing fraction curve.

Neutron-proton scattering at 90 Mev, K. Brueckner, W. Hartsough, E. Hayward, W. M. Powell (Univ. of California, Berkeley), *Phys. Rev.* **75,** 555–564 (1949). The recoil proton tracks observed in a 16-in. cloud chamber filled with hydrogen were studied during the irradiation of the chamber with a narrow beam of 90-Mev neutrons. The results show that the *n-p* scattering at these energies is not isotropic in the center of gravity, and is not symmetric about 90°. The proton peak in the forward direction indicates some exchange force.

Electron-neutrino angular correlation in the beta-decay of He^6, J. S. Allen, H. R. Paneth, A. H. Morrish (Univ. of Chicago, Ill.), *Phys. Rev.* **75,** 570–577 (1949). The recoil nuclei from He^6 decay were detected by an electron multiplier tube, and the electrons by an end-window Geiger counter. By varying the potential on grids in the path of the recoils, an energy spectrum was obtained for angles of 180° and 162° between the recoil and electron paths. Coincidence measurements assured that the particles from a single event were correlated. The best agreement with theory was obtained with the correlation function $1 - v/3c \cos\theta$, corresponding to an axial vector interaction.

Spin dependence of slow neutron scattering by deuterons, E. Fermi, L. Marshall (Univ. of Chicago, Ill.), *Phys. Rev.* **75,** 578–580 (1949). Information on the scattering lengths for the doublet and quartet states of spin orientation for the neutron and deuteron has been obtained from cross sections. For epithermal neutrons, indium and silver neutron resonances were used; for neutrons of 5.43 A.U., a filter was used. The two scattering lengths are the same in sign but may differ by a factor of two.

Mixed cosmic-ray showers at sea level, C. Y. Chao (Massachusetts Inst. of Technology, Cambridge), *Phys. Rev.* **75,** 581–590 (1949). Cloud-chamber photographs have been made of showers containing both slow and fast heavy particles as well as the electron component. The cascade showers are discussed in regard to initiating event. A star is produced in one case by a penetrating particle shower.

East-west asymmetry and latitude effect of cosmic rays at altitudes up to 33,000 feet, W. C. Barber (Univ. of California, Berkeley), *Phys. Rev.* **75,** 590–599 (1949). Using a triple-coincidence telescope the asymmetry was measured at several geomagnetic latitudes. The hard, soft, and shower-producing components were separately investigated, and the results indicate that all were produced by the same primary rays. The variation of vertical intensities of the hard and soft components with altitude and geomagnetic latitude is also given.

Relation of the cosmic radiation to sunspot magnetic moments, J. W. Broxon (Univ. of Colorado, Boulder), *Phys. Rev.* **75,** 612–619 (1949). The arithmetical sum of the magnetic moments of all visible sunspots has been statistically compared with the intensity of the cosmic radiation. Various phase relations were discovered between pulses in the two distributions.

The radioactivities of some high mass isotopes of cobalt, T. J. Parmley, B. J. Moyer, R. C. Lilly (Univ. of California, Berkeley), *Phys. Rev.* **75,** 619–623 (1949). Using neutron bombardment of nickel samples enriched in isotopes 61, 62, and 64, activities of Co^{61} (electron 1.3 Mev, no gamma, 1.75-hr half-life) and Co^{62} (electron 2.3 Mev, gamma 1.3 Mev, 13.9-min half-life) were identified.

Quantitative measurements with scintillation counters, H. Kallmann (Kaiser-Wilhelm Inst., Berlin-Dahlem, Germany), *Phys. Rev.* **75,** 623–626 (1949). The efficiency of various fluorescent materials used as scintillation counters has been measured. The efficiencies depend on the physical light yield (fraction of absorbed energy transformed into light), and the practical light yield (amount of light actually obtained for a given radiation, due to the possible opacity of the material to its fluorescent radiations). The organic phosphors (naphthalene, phenanthrene) and KBr phosphors are superior in the latter respect to the sulfide phosphors.

A determination of the ratio of the masses of π- and μ-mesons by the method of grain counting, C. M. G. Lattes, G. D. S. Occhialini, C. F. Powell (Univ. of Bristol, England), *Proc. Phys. Soc.* **61A,** 173–183

(1948). Analysis of the tracks in more than thirty events in which the secondary μ-meson (of energy about 4 Mev), produced by a π-meson at rest, comes to the end of its range in the emulsion, gives a ratio for the masses. The method of determining the mass ratio is explained, and the ratio m_π/m_μ is given as 1.62 ± 0.12. The fluctuations in the ratio indicate that the decay of a π-meson is accompanied by emission of a neutral meson of mass 115 ± 30 electron masses.

Determination of the masses of charged particles observed in the photographic plate, Y. Goldschmidt-Clermont, D. T. King, H. Muirhead, D. M. Ritson (Univ. of Bristol, England), *Proc. Phys. Soc.* **61A,** 183–194 (1948). The scattering of 40 protons and 160 mesons in photographic emulsions was investigated and the masses involved evaluated by the curvature of the path due to scattering. The proton tracks were used as a check on the method, and the value observed was 1.2 ± 0.1 proton masses (possibly due to the presence of deuterons and tritons). The results for mesons were: π-mesons, $260 \pm 30m_e$ (electron masses); μ-mesons, $205 \pm 20m_e$; σ-mesons, $275 \pm 15m_e$; ρ-mesons, $200 \pm 10m_e$.

Continuous γ-emission in neutron-proton collisions, M. Krook (Univ. of Birmingham, England), *Proc. Phys. Soc.* **62A,** 19–26 (1949). A calculation is made of the brehmsstrahlung to be expected in n-p collisions for bombarding energies up to 20 Mev, using a central exchange force model. The value of 10^{-28} to 10^{-29} cm^2 obtained for the cross section is too small to be measured at present.

Further experiments with an adjustable Geiger-Müller counter, A. G. Fenton, E. W. Fuller (Univ. of Birmingham, England), *Proc. Phys. Soc.* **62A,** 32–40 (1949). An investigation was made of the variation of multiple pulses with counter voltage, anode diameter, and total pressure of filling. Plateau curve slopes include the effect of increased counter efficiency and more multiple pulses, which may be either spontaneous or due to the effect of previous genuine pulses. The charge per pulse is found as a function of operating voltage. Using the counter as an ionization chamber showed that, in the proportional region, first log I was proportional to V (I = ionization, V = counter voltage), then log-log I was proportional to V. As the Geiger region was entered, a sharp break in the log-log I curve indicated a discharge spreading.

Investigations on the binding energy of heavy nuclei, R. Huby (Univ. of Bristol, England), *Proc. Phys. Soc.* **62A,** 62–71 (1949). Approximate calculations of the binding energy of heavy nuclei on the basis of the Møller-Rosenfeld interaction were made using first and second-order perturbation approximations on the Fermi gas model. About 50% of the binding energy is explained in this way, by the use of parameters obtained from the characteristics of light nuclei.

Trochotron design principles, G. W. Monk, G. K. Werner (Carbide and Carbon Chemicals Corp., Oak Ridge, Tenn.), *Rev. Sci. Instr.* **20,** 93–96 (1949). The perfect focusing properties of the crossed electric and magnetic fields of the trochoidal type of mass spectrometer are derived (this term describes the ion paths). Nomographs and general trochoids are given to correlate the design variables.

Design of the radiofrequency system for the 184-inch cyclotron, K. R. MacKenzie, F. H. Schmidt, J. R. Woodyard, L. F. Wouters (Univ. of California, Berkeley), *Rev. Sci. Instr.* **20,** 126–133 (1949). The f-m oscillator and associated circuits for use with the 184-in. cyclotron to give 190-Mev deuterons are described. The normal voltage delivered by the r-f at the front of the dee is 20 kv. A condenser whose capacitance is changed mechanically by rotating the plates 100 times per second sweeps the frequency from 12.5 to 9.0 megacycles per second.

Table of isotopes, G. T. Seaborg, I. Perlman (Univ. of California, Berkeley), *Revs. Mod. Phys.* **20,** 585–667 (1948). A revision of the well-known isotope tables issued in 1944, this article includes the following data on active and stable isotopes of all the elements: atomic and mass number, percent abundance, types of radiation emitted together with the corresponding energy of emission, and methods of producing active isotopes.

The metabolic properties of the fission products and actinide elements, J. G. Hamilton (Univ. of California, Berkeley), *Revs. Mod. Phys.* **20,** 718–728 (1948). Twenty-two different radioelements, both of the actinium series and fission product group, were investigated for their metabolic properties in the rat. The absorption rate, accumulation in the principal organ of retention, and rate of elimination are given. Radioautographs are shown.

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for different elements) for bombardment with X-rays from betatron electrons. Calibration was by means of the deuterium, beryllium, carbon, and nitrogen thresholds. The thresholds of various (γ,p) and (γ,n) reactions were determined, and the excitation curves (up to 22 Mev) appear similar in shape, with the exception of that for Ta. The thresholds are correlated with the packing fraction curve.

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East-west asymmetry and latitude effect of cosmic rays at altitudes up to 33,000 feet, W. C. Barber (Univ. of California, Berkeley), *Phys. Rev.* **75,** 590–599 (1949). Using a triple-coincidence telescope the asymmetry was measured at several geomagnetic latitudes. The hard, soft, and shower-producing components were separately investigated, and the results indicate that all were produced by the same primary rays. The variation of vertical intensities of the hard and soft components with altitude and geomagnetic latitude is also given.

Relation of the cosmic radiation to sunspot magnetic moments, J. W. Broxon (Univ. of Colorado, Boulder), *Phys. Rev.* **75,** 612–619 (1949). The arithmetical sum of the magnetic moments of all visible sunspots has been statistically compared with the intensity of the cosmic radiation. Various phase relations were discovered between pulses in the two distributions.

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. . . HAROLD BROWN

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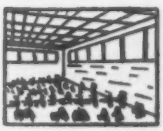

NUCLEAR INSTRUMENT HANDBOOK

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HISTORY AND MARKET ANALYSIS

Survey of Nucleonics Instrumentation Industry

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GEIGER COUNTERS and ion chambers made the front pages in the summer of 1946 but they were, at that time, an old story to physicists. Today these instruments are manufactured in many shapes and sizes by a growing number of industrial concerns. NUCLEONICS is dedicating a large section of this issue to a survey of the current status of the radiation detection instrument field. It is fitting, therefore, that this survey be prefaced by a few remarks about the industry which has arisen to produce this equipment.

We shall not discuss the non-commercial production which was carried out by many research institutes and universities. Most such researchers, including those who pioneered in the development of G-M tubes and other instruments, have given up the practice of making their own gadgets and now buy products from commercial organizations.

Early Days of Industry

The early days of this special instrument business go back to the late twenties just before the stock market crash. At that time John Victoreen, who had been in the radio business, founded the Victoreen Instrument Company for the purpose of producing instruments for the measurement of X-rays. The initial activity of the company was modest and the staff consisted of about 15 people. Instruments produced centered about the needs of the radiologist; condenser chambers or r-meters were standard products.

Except for radium and radon, radioactive materials were not in great use at that time. Actually there was very little need for instruments of high sensitivity. Only occasionally was a Geiger counter or a special electroscope pressed into service—to hunt for some radium that had been misplaced. More often than not, the hospital authorities would seek help from a nearby university and a Geiger counter would be hastily improvised as a survey meter to aid in locating the lost material.

Only a few companies (Herbach & Rademan entered the Geiger tube field about 14 years ago) specialized in building instruments, which actually were designed for the measurement of X-ray dosage. About 1930 F. M. Eck (of the present Eck & Krebs Company) began working with Columbia University on the construction of G-M tubes which were later to become essentially standard as the Eck & Krebs thin-walled glass counter tube. Cyclotron research stimulated some additional interest in nucleonics instrumentation but not enough to warrant the growth of many new companies. Only a relatively few, such as Technical Associates and Cyclotron Specialties, responded to this stimulus.

Such was the condition of the industry prior to World War II. Dollar sales were small, probably about $100,-

000 for the entire industry. Instruments were comparatively crude although a few were about as highly developed as they are today. Then came the war and the now famous Manhattan Project. There came too the demand for many new kinds of radiation instruments. They had to be more rugged, more reliable, and of higher sensitivity than anything which had been built previously.

Wartime Developments

To meet the exacting requirements, special development centers were established within the Manhattan Project, the principal ones being at the Metallurgical Laboratory (Chicago), the Clinton Laboratory (Oak Ridge) and the Los Alamos Laboratory (New Mexico). New types of instrumentation to measure alpha particles, weak beta particles, or beta particles in the presence of other intense radiation, had to be developed. Monitoring equipment had to be installed in the various laboratories of the Project. Automatic registering devices were made to "count" the contamination of a worker's hands and feet. Instruments bearing whimsical names, such as Zeus, Zeuto, Poppy, Betty Snoop and Pluto, were brought forth to fit the special needs of each site.

Before the war, neutron counters had been rarities even in research laboratories. The development of nuclear reactors required that new types of extremely reliable neutron counters be perfected. Counters were filled with boron trifluoride or lined with a boron compound. Then, as the separation plants put out a supply of B^{10}, the more useful isotope of boron, a compound containing enriched boron was used in the counters.

In the same way the research problems and health hazards encountered in dealing with plutonium created a demand for reliable alpha particle detectors. Even today the ideal alpha survey instrument is far from a reality, and in the war years the type of instrument then in vogue performed satisfactorily only if carefully serviced.

Pulse analyzers were in demand by the radiochemists. But by far the greatest demand was for a reliable scaling circuit. A series of different types was evolved and finally the Project almost standardized upon the Higinbotham circuit (scale-of-64), which finds much favor, even today.

There were, of course, many instruments designed for specific process control. Still others were designed for use in association with the piles built at Oak Ridge and Hanford. Other varieties found application in the vastness of the separation plants at Oak Ridge.

There was a marked concentration upon the development of devices for the measurement of low currents. The old standby, the FP-54, was used but, because of its disadvantages for many applications, other approaches to the problem of small current measurement were made. Lindemann electrometers, which had served cosmic-ray physicists so well, were used for some applications. Since the supply of these quartz fiber instruments was very limited, the Metallurgical Laboratory set up a quartz shop and produced over 100 of a redesigned electrometer. The Lauritsen electrometer, as adapted to the pencil-type personnel meter, proved to be extremely useful for many operations. Difficulties encountered in the erratic behavior of the usual type of insulators led to the development of specially molded high resistance plastics, such as polyethylene. Other approaches to the low-current problem concentrated on the use of new vacuum tube electrometers, and one very useful approach employed the vibrating reed electrometer.

The task of equipping the individual laboratories of the Manhattan Project

was a formidable one. It was not accomplished without some grief but the job was done. The health protection record of the Project clearly proves that the job was well done. Since there were so few companies to which work could be allocated, the laboratories often did the jobs themselves using crude production set-ups to turn out small quantities of semi-standardized equipment. A few companies were put under contract with the Manhattan Engineer District. Small electrometer tubes, such as the Victoreen subminiature style tube, were in great demand and special provisions were made for producing this critical item.

Project Crises

There were three distinct instrument crises during the course of the war. There was the highly secret OPERATION PEPPERMINT, which was started when it was thought the Germans might use radioactive materials against our invasion forces. For this the multirange Model 247 Victoreen ionization chamber instrument was developed and produced in quantity. This was probably the first standard survey meter of its type. Fortunately, no radioactive agents were used on the Normandy beachheads and the instruments were not needed. Had it been necessary to use them, it is interesting to speculate on the value of using any instruments with untrained personnel in combating a new hazard of warfare.

Another crisis was the outfitting of the Hanford Engineer Works (Washington) with the equipment needed to monitor plant activities. Much of this task was assigned to the Metallurgical Laboratory, which had been working in close cooperation with duPont, the company which ran Hanford. The laboratory itself was unable to mass produce enough instruments and subcontractors, such as Hallicrafters, were called in. The National Technical Laboratories also produced instruments for the Hanford plant.

The last wartime push in instrumentation was created by the Trinity bomb test at Alamogordo. This should have served as a proving ground for survey instruments to be used in future tests, but it did not.

Post-war Boom

With the ending of the war, the industry underwent a boom. New companies were formed by technical personnel who left the Manhattan Project. Typical of these new enterprises are Tracerlab and Atomic Instrument Company in the East, Nuclear Instrument and Chemical Corp. and Radiation Counter Laboratories in the Chicago area, and Berkeley Scientific Company in the West. All of these organizations, and a few others, started out as modest efforts. Some marketed a modified version of the Higinbotham scaler. Others put out a line of G-M tubes. In general all profited from the extensive investigations carried on during the war; few put much of their own money into new research and development.

Radioisotopes became available from the Oak Ridge pile and things started to click in the rejuvenated industry. The small companies went through their growing pains and began to prosper. Meanwhile, big business which had been marking time before plunging into the field, began to take an interest but decided to wait it out a little longer.

About this time plans were rushed through to completion for the bomb tests in the Pacific. The Army called upon the Victoreen Instrument Company to supply the bulk of its requirements. OPERATION CROSSROADS was carried out in the Bikini lagoon with Col. Stafford L. Warren as Chief Radiological Safety Officer. Not one person out of the 42,000 present was injured by the radioactive hazards at Bikini, even though they were more

severe than most had anticipated. Most significant was the fact that the Navy was left with some of its target fleet contaminated with radioactive materials. Ships had to be evaluated as to damage and some of them had to be decontaminated. Instruments were borrowed from the Army, but, thereafter, the Navy undertook to develop its own set of measurement equipment.

At the same time that the Navy undertook its instrument program, the Atomic Energy Commission was emerging from the realm of Washington paper work and becoming a reality. The AEC immediately sponsored an enlargement of the radioisotope program and realized that it would also have to encourage new companies to produce instruments for research involving radioactivity. Consequently, it actively did this at a time when the Navy was doing the same thing. There was little evidence of any coordination between the two agencies. In general, the Navy went to big business with its problem whereas the AEC looked to smaller companies for assistance. More and more money began to be spent for development rather than for production and this was an exigency for which some of the interested companies were poorly qualified. They were better suited to production.

The Market

Some impetus to the industry was given by the surprisingly great use of radioisotopes. Larger companies, including National Technical Laboratories and Kelley-Koett, actively entered the radiation field with a line of instruments. As things began to stabilize, manufacturers found themselves in a competitive field. Some standardization was taking place within the AEC and orders were placed on a bid basis for quantities of instruments. Prices dropped. Then too, things were not eased by orders which industry had been led to expect would be placed by the military. Thus 1947 saw the temporary stabilization of the industry with the supply exceeding the demand. The impetus of the Eniwetok bomb tests did little more than give a few companies a shot-in-the-arm. It was clearly recognized that there were too many companies in the business. Competition for the existing market would either eliminate some of the smaller concerns or it might conceivably convince big business that it could afford to disregard the field.

In spite of the ominous signs, at least six new companies devoted to the development and production of these instruments were organized in 1948. Large military orders were still only a rumbling in Washington. It was known that some companies had research and development contracts with the military but there was no evidence of any production orders. Only recently have military orders for production lots of instruments been received by instrument companies and indications are that the total dollar value of these is considerably less than one million dollars. Today this is only a small fraction of the output of the industry.

At this point it is of interest to examine the types of instrument buyers other than military. The market may be broken down as follows:

1. Atomic Energy Commission
2. Research institutions and universities
3. Government agencies other than AEC
4. Industry
5. Prospectors

Of these markets, the industrial one seems to be the least tapped, for strangely enough, industry has been extremely sluggish in applying the fruits of atomic energy research. This is no longer explained by a scarcity of supply as only a small fraction of the

AEC's radioisotopes has been going to industry. With the AEC encouraging prospecting for uranium deposits, a considerable number of survey-type instruments has been sold to prospectors.

Prior to the war the dollar value of the yearly sales was probably of the order of $100,000. No accurate figures are available for the period from 1940-1945 but the total must have been more than $10,000,000. The post-war period represents the significant one and more precise figures have been made available for this survey.* On the basis of the data supplied and assumed data for the few companies which did not provide sales data, the following figures have been calculated:

Year	*Yearly sales of instruments*
1946	$ 800,000
1947	$1,700,000
1948	$3,500,000
1949 (Est.)	$4,500,000

The sales curve should continue rising for at least five years. Thereafter, its rise or fall will, in large measure, depend upon the over-all usefulness of radioisotopes to science and industry. The Atomic Energy Commission will continue to be a big consumer but unless the present status is upset by unexpected developments the AEC's demands should not continue to rise.

The purely civilian market, i.e., universities, institutes and industry, should remain fairly steady and a sharp increase would seem unlikely. Radioisotopes are useful but not dominantly important tools and their use seems definitely to have limits. Another phase of the civilian market is the possible stockpiling of survey instruments for atomic disaster purposes. A recent report by the Office of Civil Defense Planning might lead one to believe that large numbers of instruments would be procured for this purpose, but there is no time scale indicated for the purchase. It would seem highly desirable to predicate quantity procurement upon the successful small scale employment of the instruments by token civilian task forces and upon a realistic appraisal of the probable hazards to be encountered. Many believe a program of education to be more fundamental than a program of procurement. If every large city is to have a stock of radioactivity measurement instruments on hand and if present type instruments are procured, we will be in a constant state of jitters every time a Geiger counter misbehaves.

While the industry is still in its infancy, there is no better time to think of standardization. Today not even the terminology is standard, much less the instrumentation. Earlier this year the Radio Manufacturers' Association and the Institute of Radio Engineers held an exploratory meeting to consider the problems of standardizing certain nuclear measurement equipment. Progress in this direction is definitely indicated; the rate will depend upon the degree to which industry cooperates. Today many of the instruments produced vary considerably in reliability. Mindful of the truism that "a bad instrument is worse than none at all" it will pay for the industrialists to meet commonly agreed upon levels of performance, for an instrument used for health protection must be a *good* instrument.

* The author wishes to thank those manufacturers who cooperated so splendidly by making available to him financial data of a confidential nature.

MEASUREMENT PROBLEMS

Nuclear Physics

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Gamma Rays

Geiger-Müller counters must be operated in connection with stabilized power supplies, electronic amplifiers, and output circuits.

Light-element cathodes. The common forms of these include the use of silver deposits on the inside of glass envelopes, graphite, or copper as cathodes. In the energy range from low to 3 Mev, the detection efficiency of this type is small at low energy, reaching less than 0.5% at high energy.

Heavy-element cathodes. These counters can be made with lead, platinum or bismuth cathodes. They have relatively high efficiencies of 0.2% up to 1 Mev, and increase in efficiency at higher energies as do counters with other cathodes.

Cell-type construction. Counters built with internal fins, plates, baffles, or banks of cylindrical cathodes, all within the same envelope, can increase the detection efficiency as much as a factor of ten over conventional construction.

Scintillation counters used in conjunction with photomultipliers such as a 931-A or 1P21, and associated electronic amplifiers.

Types of crystals which fluoresce under ionizing radiation and are transparent to their own radiation are satisfactory. They exhibit very high efficiencies, from 60% to almost 100%.

Crystal counters are operated with high-gain amplifiers to detect the ion current in the crystal lattice.

Room temperature. Selected diamond and cadmium sulfide crystals operate at room temperature. Very high efficiency, close to 100%.

Liquid air temperatures. Silver chloride type of crystal operates successfully at liquid air temperatures. Detection efficiencies close to 100%.

Photographic emulsions

Autoradiographs. Contact detection using commercial types of X-ray film.

Deuterium loaded emulsions with D_2O may be used by detecting the photodisintegration of deuterium above 1 Mev gamma rays.

Beta Rays

Geiger-Müller counters must be operated in connection with stabilized power supplies, electronic amplifiers and output circuits.

Thin-walled counters of glass or thin metal are satisfactory for high energy beta rays; their efficiency is close to 100%.

End-window construction allows for detection of wide range in energy. Large unsupported mica windows may be used to less than 0.1 Mev. Collodion or Zapon windows when supported on grids may be used for detection of radiation down to 2 or 3 kev.

No windows used when source may be placed within the counter, either in the gas with flowing gas or sealed off. When activity is present in the gas phase, the efficiency can reach 100%.

Proportional counters must be operated in connection with stabilized power supplies, electronic amplifiers, pulse discriminators, and output circuits. Constructional details are similar to those outlined under Geiger-Müller counters. Since output pulse from the counter is proportional to the beta-ray ionization, discrimination of the radiation ionization may be accomplished.

Electron multipliers. Special multipliers built with plates of metal such as beryllium-copper have electron efficiencies of 100% below 0.5 kev, the efficiency decreasing to about 40% at 6 kev.

Photographic emulsions

Autoradiograph by direct contact using films such as Eastman No-Screen.

Direct detection of tracks in nuclear emulsions.

Recording instruments such as spectrometers which can be operated completely in a vacuum.

Cloud chambers are used with control and expansion circuits. For reasonable efficiency, typical conditions would be four atmospheres pressure with isopropyl alcohol and water.

Ionization chambers have a high efficiency obtainable with high pressure. Typical gas filling would be five atmospheres of argon with 5% CO_2. Using electron collection, one can obtain high resolving times. Chamber must be used in connection with an electrometer.

Electrometer tubes, such as FP-54, give electrical output directly.

Mechanical electrometers, such as moving fiber electrometers, give visual or photographic recording, and vibrating reed electrometers give electrical output.

X-rays

Geiger-Müller counters must be operated in connection with stabilized power supplies, electronic amplifiers, and output circuits.

End-window counters allow the passage of radiation. Windows of mica, glass, or plastic. With appropriate knowledge of critical absorption limits, windows may be arranged to pass X-rays but absorb beta rays.

Proportional counters must be operated in connection with stabilized power supplies, electronic amplifiers, pulse discriminators, and output circuits.

End-window counters have similar characteristics to Geiger-Müller counters except that the pulse size is proportional to the ionization produced in the counter.

Photographic film

Direct detection by exposure to the radiation.

Recording instruments such as crystal spectrometers.

Ionization chambers used in connection with moving fiber electrometers, or current amplifiers.

Thimble chambers with carbon walls give accurate readings in roentgens.

Electron multipliers have shorter resolving times but are not as efficient for detection of about 1 A.U. X-radiation as a Geiger-Müller counter.

Neutrons

Geiger-Müller counters operated in connection with stabilized power supplies, electronic amplifiers and output circuits.

Foils which become radioactive when bombarded by neutrons are measured with Geiger-Müller counters.

Proportional counters operated in con-

nection with stabilized power supplies, electronic amplifiers, pulse discriminators and output circuits.

BF_3 filled counters detect slow neutrons by the neutron disintegration of boron, producing alpha rays as the ionizing particles. BF_3 enriched in B^{10} increases the efficiency and allows detection to several Mev. Surrounded by a thick paraffin shell, the detector can have uniform sensitivity from 10 kev to 3 Mev.

Proton recoils can be used as the ionizing medium. Tristearin, polythene, or other hydrogeneous films mounted as radiators in the counter can lead to efficiencies of 1% for 1 Mev neutrons. Counters filled with hydrogen gas also provide a source of proton recoils.

Helium recoils have also been used to detect neutrons in the 1 Mev range using proportional counters with high pressure helium or helium-argon mixtures to several atmospheres.

Photographic emulsions are effective by the use of the proton recoils from the hydrogeneous materials in the photographic films.

Ionization chambers are used in connection with linear amplifiers or vibrating reed electrometers and associated circuits.

BF_3 filled chamber with several atmospheres of B^{10} enriched gas. To detect no neutrons above 0.5 ev, it can be surrounded by a thick cadmium filter. Efficiency for slow neutrons is around 1 to 5%.

Proton recoils can be obtained by filling chamber with H_2. For the 1-3 Mev range, typical filling is 2 atmospheres of H_2 and 4 atmospheres of A.

U^{238} fission for slow neutron detection can be measured in 96% A and 4% CO_2 mixture, with electron collection for rapid response.

U^{238} fission for fast neutron detection, uranium-coated chamber surrounded by cadmium to avoid thermal neutron fission of U^{235}.

Bismuth plates in chamber to give fission particles, for neutrons above 50 Mev, efficiency about 10^{-6}.

Alpha Rays, Protons and Deuterons

Proportional counters operated in connection with stabilized power supplies, electronic amplifiers, pulse discriminators and output circuits. Can separate alphas, protons or deuterons from intense beta-ray and gamma-ray backgrounds. To avoid recoil protons in measurement of protons, no hydrogeneous material must be exposed to the sensitive volume.

Photographic film. Autoradiographic detection is 100% efficient.

Ionization chambers can be run with d-c amplifiers, linear amplifiers, vibrating reed or fiber electrometers. Efficiency can be 100% and can be insensitive to beta rays or gamma rays.

Electron multiplier with beryllium-copper plates is 100% efficient, and, since it operates in a vacuum, it can be used for very low energy alpha rays.

Scintillation counters used with photomultiplier tubes and their associated apparatus.

ZnS crystals have high efficiency for alpha particles but give no effect with beta rays or gamma rays.

Anthracene and similar crystals that are transparent to their own fluorescence have high alpha-ray efficiency but are also sensitive to beta rays and gamma rays.

Cloud **chamber** needs control circuits and expansion mechanism, and can be made sensitive to heavily ionizing particles but not to beta rays and gamma rays.

Radiobiology

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THE GENERAL FIELD of radiobiology may, for present purposes, be divided into (1) tracer studies with radioactive isotopes and (2) the biological effects of high energy radiations. The problems of instrumentation and measurement are quite different for the two fields and will be considered separately.

In biological tracing experiments care must be taken to keep the amounts of administered radioactive isotopes small so that normal physiological function is disturbed as little as possible by the radiation. Since, after the isotope has been distributed through the tissues, the amount of active material in a small sample taken for assay may be very low, the usual problem in tracer studies is the accurate measurement of relatively low activities. Frequently measurements must be made on samples which give a response about equal to that from the normal background radiation. This sets the lower level of sensitivity for an assay by any method: hence laboratory contamination must be kept well below that required from a health standpoint.

Radioactive isotopes are usually assayed by determination of the beta or gamma activity of a sample placed in a fixed geometrical relation to the detecting device. Only occasionally is it necessary to trace a pure alpha emitter. Whenever possible the assay is made by measurement of the beta activity, which can be detected with considerably greater efficiency than the gamma activity.

In the selection of a suitable assay method the first choice must be made between Geiger-Müller counters and ionization chambers connected to some form of electrometer. In general, G-M counters show a greater sensitivity than ionization chambers and hence are more widely used. However, with low energy radiations the ionization chamber comes into its own and may prove to be the instrument of choice.

Many isotopes have been used in biological investigations but, of these, only the few listed in Table 1 have been used to any great extent.

H^3, C^{14} and S^{35} are of great biological interest but it will be noted from Table 1 that none of these emit gamma radia-

TABLE 1

Radioactive Isotopes of Biological Importance

Isotope	*Half-life*	*Beta energy (Mev)*	*Gamma energy (Mev)*
H^3	31 y	0.015	None
C^{14}	5100 y	0.15	None
Na^{24}	14.8 h	1.4	1.38 2.76
P^{32}	14.3 d	1.72	None
S^{35}	87.1 d	0.14	None
Fe^{59}	47 d	0.26 0.46	1.1 1.3
I^{131}	8.0 d	0.6	0.37 0.08

TABLE 2

Counter-type Instruments

Type	*Most useful for*	*Advantages*	*Disadvantages*	*Ref.*
Internal	H^3, C^{14}, S^{35}	No absorbing window; can count gas samples	Refilling required; reproducibility requires care; some gases not suitable	*1, 2, 3, 4*
Flow Gas	C^{14}, S^{35}	No absorbing window; reproducible filling	Not suitable for gas samples	*2*
Thin Window (< 5 mg/cm^2)	C^{14}, S^{35}	Filling constant	Some window absorption	*5*
Standard ~ 20 mg/cm^2	Na^{24}, P^{32}, I^{131}	Filling constant	Insensitive to low energies	
Gamma	Any γ-emitter	Measurements from intact subject	Low efficiency	
Crystal	Any	High γ-efficiency; short resolving time	Small size	*6*
Scintillation	Any	High γ-efficiency; short resolving time	High background current	*7, 8*

tion and that the betas are of relatively low energy. It is difficult to get windows thin enough to obtain a high counting efficiency with the low energy beta radiations from these three isotopes. Consequently ionization chamber techniques have proved quite useful in studies of these particular materials.

In Table 2 are listed some of the general types of counter instruments, together with pertinent data concerning them. The types of counters listed here will take care of the majority of counting problems for which Geiger counters are suitable. In a few cases special instruments must be developed for the problem in hand. Such a special problem is the determination of Fe^{55} and Fe^{59}. To separate these activities it has been found advantageous (*9*) to use a special counter tube with a beryllium window to detect the soft X-rays emitted as a result of the *K*-capture by the Fe^{55} isotope.

Any of the Geiger-Müller counters listed in Table 2 may be connected either to a scaling circuit and a mechanical register or to a rate meter with a continuous recording device. Both of these methods have their advocates, but, in general, it seems that, for biological assay of individual samples, the scaling circuit has some advantages, whereas for determination of decay curves over relatively long periods of time the rate meter is superior. In most cases it is desirable to obtain a decay curve of the isotopic sample being used since it is not always certain that the sample is pure and that the simple logarithmic decay anticipated from a single isotope is actually being obtained.

Table 3 lists some of the types of electrometers which can be used with ionization chambers for radioactive assay. These detecting devices can be used to determine any type of radiation, the exact type depending upon the design of the ionization chamber. In biological assay work the ionization chamber is most useful for determining alpha particles and low energy beta

TABLE 3

Ionization Chamber Instruments

Type	*Advantages*	*Disadvantages*	*Ref.*
Lauritsen electroscope	Simple, rugged, reliable	Measurements tedious at low levels	*9*
Vacuum tube electrometer	Rugged, simpler than most electrometers	Some circuit drift at high sensitivity	*10, 11, 12*
Synchronous condenser	A-C amplification, low drift	Expensive	*13*
Lindemann	Small, simple, cheap, not sensitive to level of activity	Measurements tedious at low levels	*14*

particles and it is assumed that the ionization chambers will be designed in accordance with these demands. Many types of electrometers have been devised and there is not space here to discuss all of them. Table 3 lists only those which have proved to be most useful in biological work.

In determining the biological effects of high energy radiations it is customary to radiate either whole animals or simple chemicals or biological systems with the radiation of choice, which may be any of the nuclear radiations or X-rays. The problem here is not one of obtaining high sensitivity because, in general, the radiation rates are relatively high. Rather it becomes a matter of measuring high intensities and determining the fraction of the radiation absorbed by the system irradiated. Biological materials may be irradiated either with X-rays, gamma rays or neutrons from external sources or with alpha or beta particles from radioactive materials introduced internally. The radioactive materials can be standardized with one of the appropriate instruments listed in Tables 2 and 3. In general rather large doses of radiation are required to produce observable biological effects and the assay will not require the determination of small quantities. Rather, dilution may be necessary to bring the activity within range of the counting equipment. From a knowledge of the energy of the radiations and the concentration of the administered material the dose can be determined (*15, 16*).

Ionization chamber instruments are used almost exclusively for measuring X- and gamma-ray doses since with the proper choice of chambers the response will be proportional to the dose received by the tissue. Dose rates may be as high as 100 r per hour and care must be taken to insure that the ion chamber is saturated at the highest rate and the wall thickness of the chambers must be chosen so that there will be electronic equilibrium in the gas space.

For use with X-rays having energies below 100 kev a chamber with nylon walls about 0.005 in. thick may be used in connection with an r-meter. These chambers are not air equivalent at low X-ray energies (*17*) and require large corrections below 20 kev but are the only chambers now available for low quantum energy measurements.

For quantum energies between 70 kev and 200 kev, bakelite wall chambers are customarily used and yield readings practically independent of energy. These chambers are quite satisfactory for energies up to 1.5 Mev and may be calibrated with the gamma rays from radium which have an effective energy of about 1.3 Mev.

Measuring techniques for high energy

X-rays (20-100 Mev), such as are obtained from the betatron, are not well established. Lead-jacketed ionization chambers have been used (*18, 19*) but it is doubtful if they are air equivalent. Much work remains to be done in this energy range.

In the early biological experiments with neutrons standard bakelite chambers for 200 kev X-rays were used in lieu of better instruments (*20*). At the present time hydrogen-filled counter tubes appear to be most suitable for measurements of fast neutrons. Slow neutrons are usually measured with a boron-filled counter (*21*). If the velocity distribution of the neutron beam is known the induced radioactivity of thin foils can be used to measure neutron intensities. Although relative values of neutron fluxes can be readily measured there is some doubt as to the dose received by the material being irradiated and improved techniques are needed.

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Health Physics

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THE FOLLOWING PAGES contain a tabulated summary of measurements of interest in the field of health physics. Being a summary, the entries must of necessity be brief. The reader is advised to consult detailed articles that discuss the various components. Some of these references are given on p. 113.

Column 3 in the tabulation lists the "Approximate Allowed Limits of Intensity, Dose or Contamination." These have at times, been referred to as "tolerances." Actually the health physicist regards no contamination or radiation other than natural background as being "tolerable." Practical

considerations, however, have required that some limits be set, and those listed in column 3 indicate the orders of magnitude adopted in practice. They are tentative limits, and are constantly undergoing revision. Most of the figures given are chosen from the first three references listed in the bibliography.

The dose units commonly used are the roentgen (r), the roentgen-equivalent-physical (rep), and the roentgen-equivalent-man (rem). The roentgen, used only in connection with X- or gamma rays, represents that quantity of electromagnetic radiation which will cause the energy absorption of 83 ergs per gram of air. This is approximately equivalent to 83 ergs per gram or cc of tissue. The rep is used in connection with all types of radiation, electromagnetic or particulate. It represents that quantity of any radiation that will cause the absorption of 83 ergs per gram or cc of tissue.

Equal quantities of energy absorbed from different types of radiation may cause unequal biological effects. For this reason the rem is introduced, to reduce dosage to a common denominator, the relative biological effectiveness. One rem represents that quantity of any radiation which will produce the same biological effect as one roentgen of electromagnetic radiation (X- or gamma rays).

A table of equivalent units used in some laboratories follows:

X- and gamma rays	1 rem = 1 rep = 1 r
beta rays	1 rem = 1 rep
alpha particles	1 rem = 0.1 rep
thermal neutrons	1 rem = 0.25 rep
fast neutrons	1 rem = 0.20 rep

At the present time consideration is being given to the downward revision of the prevalent "tolerance," or maximum permissible dose, of 100 millirem per day. Suggestions have been made to reduce this figure to 20 mrem per day. Also being considered is 300 mrem/wk.

Various fluxes of thermal neutrons, from 1,250 to 4,700 neutrons per cm^2 per second for 8 hours, have been suggested as being equivalent to 100 mrem per day.

In the light of recent developments, the value for the biological effectiveness of fast neutrons has been revised upwards. Suggestions have been made to change the 1 rep equals 5 rem figure to as high as 1 rep equals 25 rem. In the past, a flux of about 200 fast neutrons per cm^2 per second for eight hours was considered as equal to 100 mrem. The new figures may be as low as 40 neutrons per cm^2 per second, and if the 100 mrem per day limit is reduced, even 40 per second will be high.

* * *

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Nature of measurement	*Type of radiation*	*Approximate allowed limits of intensity, dose or contamination*	*Measurement device suggested*
I. MONITORING PERSONNEL			
***A.* Integrated dose of incident radiation** (These devices are worn on the clothing or on the part of the body that comes closest to the radiation source.)	Beta, Gamma	500 mrep/wk	Sensitive X-ray film worn in badge or holder
			Less sensitive (insensitive) X-ray film worn in holder or badge
	Gamma	100 mr/8 hr day	Pocket ionization chamber
			Dosimeter
			Signalling pocket ionization chamber
	Thermal neutrons	100 mrem/8 hr day (1250–4700 neutrons/cm²/sec for 8 hr)	Boron-lined pocket ionization chamber
			Fine-grain nuclear film
	Fast neutrons	500 mrem/wk (*see introductory comment for flux*)	
***B.* Integrated dose of incident radiation in the vicinity of worker** (Devices not worn on person.)	Gamma	100 mr/8 hr day	Battery-operated, integrating-type ionization chamber

Approximate range of measurement device	*Indicating method*	*Comments*
30–3,000 mrep	Density read on a densitometer (density range 0–3)	Films usually processed once a week. Calibration gives mrep vs density. Part of film is covered with metal to shield against soft radiations. Reading beneath shield essentially independent of energy. "Open-window" reading energy-dependent below about 300 kev X-rays
500–30,000 mrep	"	Same as above. Used to monitor extreme exposures. Finger rings and wrist badges loaded with sensitive and insensitive film are used to monitor hand and arm exposures
0–200 mr	Read on calibrated string electrometer	Chamber is charged, then worn in the field. Read daily on charger-reader. Two chambers worn side by side to minimize errors. Energy dependent below about 300 kev X-rays. Beta rays above about 1 Mev may penetrate chamber
0–200 mr	Quartz fiber electroscope built into chamber itself	Only a charging and zeroing device needed in conjunction with dosimeter. Other characteristics similar to above meter
50 mr	Audible signal	Signal sounds when integrated dose reaches 50 mr. Small enough to carry in pocket. Convenient warning system
0–200 mrem	Calibrated string electrometer	Boron captures neutrons [$B^{10}(n,\alpha)Li^7$]. These alphas cause discharge of chamber. Read daily on charger-reader. Low gamma sensitivity. Dosimeter type also available
Sub-tolerance dose detectable	Tracks counted with high power microscope	Developmental work still in progress. Films usually processed weekly. Tracks are caused by reaction of neutron with nitrogen of film. $N^{14}(n,p)C^{14}$. Low gamma sensitivity. Another type film, which uses crossed cadmium and lead strips, is also being used. Blackening caused by (n,γ) reaction with cadmium is basis of this calibration
"	"	Developmental work still in progress. Films developed weekly. Tracks are caused by proton recoils of hydrogen of film. Cadmium shield, covering part of film, serves to differentiate between thermal and fast neutrons
200 mr	Direct reading electrostatic voltmeter	May be discharged by inverting. Ion chamber is plug-in type. (Proteximeter made by Victoreen Instrument Co.)

Nature of measurement	*Type of radiation*	*Approximate allowed limits of intensity, dose or contamination*	*Measurement device suggested*
C. Checking personnel for contamination	Alpha	~ 100 dpm per hand ~ 1000 dpm per shoe sole ~ 100 dpm per small area of clothing	Proportional counter, air-filled, large nylon window
1. Hands, shoes, and clothing			Ionization chamber, air-filled, large nylon window
	Beta, Gamma	500 cpm per hand 5000 cpm per shoe sole	GM tube (30 mg/cm² wall)
		1000 cpm for inside of shoe	GM tube (30 mg/cm² wall) mounted in a probe
2. Nasal smears	Alpha	0 dpm above background	Parallel-plate ionization chamber or proportional counter
	Beta, Gamma	Depends on isotope	Mica end-window GM tube
***D.* Monitoring radioiodine ingestion by checking thyroid gland externally**	Gamma	800 cpm	GM tube mounted in fixture
***E.* Radon content of breath**	Alpha	1 $\mu\mu$c/liter of exhaled breath (10^{-12} curie/cc)	Low background proportional counter. Dried breath sample is counting gas
***F.* Determining radioactive content of urine or blood**	Alpha	Depends on element considered	Parallel-plate ionization chamber or proportional counter
	Beta, Gamma	Depends on element considered	End-window GM counter
II. AIR MONITORING			
***A.* Dust-borne radioactivity**	Alpha	In the order of 10^{-11} μc/cc of air	Electrostatic precipitator-air mover combination. Counting done in air proportional counter
1. Spot-checking			

Approximate range of measurement device	*Indicating method*	*Comments*
From about 10 dpm per cm^2 to 20,000 dpm. Other models up to 500,000 dpm	Count-rate meter or loudspeaker	"Poppy" probe is about 1 in. × 10 in., and can be used to scan surfaces of hands or clothing. Immediate response. Large "sandwich" type chambers to monitor both sides of hand simultaneously are available. These have multiple wires and are not portable. A-C operated
One model—4,000 and 40,000 dpm full scale. Another reads about 5×10^6 dpm full scale	Calibrated microammeter	More portable than proportional counter above, but 10-sec time constant is a disadvantage
Background—20,000 cpm per tube	Scaler plus electromagnetic register, or count-rate meter	Simple, single GM tube instrument, or elaborate 10 tube, 5 channel hand and shoe monitor are available. Latter monitors backs and palms of hands, and shoe soles simultaneously
Background—20,000 cpm	"	Probe arrangement allows checking the inside of shoes for contamination that may have soaked in. Creases, flaps and cuffs of clothing may also be scanned
One model—background to 3,500 cpm. Others usually to about 10,000 or 20,000 cpm	Scaler plus electromagnetic register	Proportional counter can count alphas in the presence of high beta and gamma activity. Ionization chamber cannot do so. Smear is taken with a cotton swab and transferred to a counting plaque
Background—20,000 cpm	"	Presence of any activity in smear should encourage use of masks
Background—20,000 cpm	"	Fixture is arranged so that GM tube fits under the chin, near the thyroid
With sufficiently long count sub-tolerance quantities may be determined	"	This tolerance concentration represents about 0.1 microgram of radium fixed in the body, which is the maximum allowed quantity of this element. This assumes that 50% of the radon given off by the radium escapes through the expired air
Long counts allow determination of sub-tolerance quantities	"	Spectrographic methods are also used in this connection. For counting alphas, the sample must be dried
Background—20,000 cpm	"	Specimen is usually evaporated and residue counted. A dipping GM tube, however, may be used for urine samples in some cases
Subtolerance quantities determinable by long count	"	Dust collected on aluminum foil. Entire foil counted in specially built chamber. Correction for natural air activity made by counting after collection, and 24 hours after collection

Nature of measurement	*Type of radiation*	*Approximate allowed limits of intensity, dose or contamination*	*Measurement device suggested*
	Beta, Gamma	Depends on contaminant. In the order of 10^{-7} μc/cc of air	Electrostatic precipitator and air mover combination. Counting done in ionization chamber
			Air-mover filter paper combination. Counted with mica end-window GM counter
2. Constant-operation air-flow monitor	Alpha	In the order of 10^{-11} μc/cc of air	Combination of moving filter paper, air mover, and aluminum window scintillation counter
	Beta, Gamma	Depends on contaminant. About 10^{-7} μc/cc of air	Filter paper (fixed) surrounding a GM tube (30 mg/cm² wall), and an air-mover
3. Studies of ventilating system filters	Beta, Gamma		X-ray film placed in contact with filters. Radioautographs made
***B.* Radioactive gases not adsorbed on dust particles** (spot-check methods)	Beta, Gamma	In the order of 10^{-7} μc/cc of air	Proportional counter filled with air sample or with separated contaminant and a counting gas
			Evacuated can, filled with air sample. Sample counted in gas pig with GM tube (30 mg/cm² wall)
***C.* Constant-operation background monitor** (non-airflow type)	Beta, Gamma	Natural background usually 35–50 cpm with this apparatus	Unshielded GM tube (30 mg/cm² wall)
		Natural background is usually 0.2 to 0.3 roentgen per day	Ionization chamber and vibrating reed electrometer
			Ionization chamber, charged and left in the field
III. Monitoring Water and Sewage			
***A.* Water**	Alpha	Depends on contaminant. In the order of 10^{-8} μc/cc of water	Parallel-plate proportional counter or ionization chamber

Approximate range of measurement device	*Indicating method*	*Comments*
Tolerance quantities detectable	Scaler plus electromagnetic register	Efficiency of collection approaches 100% at an air flow rate of 3 cfm. Entire foil is counted in specially built chamber. Background activity may be corrected for by counting several times with several hours between counts
Background to about 20,000 cpm	"	High-grade paper necessary, which will not clog up, but which will collect activity efficiently. Flow rate about 10 cfm for ½ hour. Section of filter paper may be counted, if convenient, and geometry correction applied
Variations in natural background detectable	Scaler plus printing register	Still in developmental stage. Filter paper moves past air mover and counter at a constant rate
Activity builds up to equilibrium. Then variations in background are detected	Counting-rate meter and moving-chart recorder	Paper is apt to clog and miss some changes in background. New arrangement uses moving filter paper. Still in developmental stage
Individual contaminated particles may be detected		Useful technique for determining presence of minute quantities of contaminant in circulating air
Tolerance quantities detectable	Scaler plus electromagnetic register	Where possible, the contaminant is separated out and acts as part of the counting gas. This method is still being developed. May be excellent method for detecting soft radiations
Background to about 20,000 cpm	Scaler plus electromagnetic register	Sample is transferred to evacuated pig. Sample is counted when equilibrium pressure is reached
Background variations	Scaler plus printing register or count-rate meter and moving-chart recorder	GM tube may be kept outdoors, and is then protected with thin plastic sheeting
Background variations	Moving-chart recorder	Very sensitive quantitative device
Background variations	Read on calibrated string electrometer	Overgrown pocket ionization chambers of about ½ to 7 liters volume. Read in same way as pocket meters. Utility is limited by electrical leakage of insulators
One model—background to about 3,500 cpm; another model to 20,000 cpm	Scaler plus electromagnetic register	Large volumes of water are filtered, then evaporated. Residue is counted on a plaque or small watchglass. Self-absorption usually is important factor in these determinations

Nature of measurement	*Type of radiation*	*Approximate allowed limits of intensity, dose or contamination*	*Measurement device suggested*
	Beta, Gamma	Depends on contaminant. In the order of 10^{-4} μc/cc water	Mica end-window GM counter
			Dipping GM tube, or jacketed GM tube
B. Sewage	Alpha	May be same as water. Depends on contaminant and upon drainage, locale, water table, etc.	Parallel-plate ionization chamber or proportional counter
	Beta, Gamma	"	Mica end-window GM counter
IV. LABORATORY SURVEYING			
***A.* General spot-checking of shields, sources, outgoing isotope containers, etc. Monitoring external radiation hazards** (These are portable instruments.)	Alpha	Not an external hazard unless personnel work within a few centimeters of source. Accelerated alphas from high-voltage machines are more dangerous. 12.5 mrem/hr for 8 hr is permissible	Air ionization chamber, with large nylon window
	Beta, Gamma	12.5 mrep/hr for 8 hr	"Air-wall" ionization chamber survey meters
			GM tube mounted in probe or in instrument itself. 30 mg/cm² tubes as well as mica end-window GM tubes are used
			Lauritsen electroscope

Approximate range of measurement device	*Indicating method*	*Comments*
Background—20,000 cpm	Scaler plus electromagnetic register	Same as for alpha
Background—20,000 cpm	Scaler plus electromagnetic register, or count-rate meter and moving-chart recorder	Dipping tube may be used in flowing system. May not detect as low activity as the evaporation technique, but is more convenient in some cases. Small liquid samples may be poured into jacketed tube. This may not be as sensitive as evaporation technique either, but is necessary when activity is volatile
Various models available. Background to 3,500 or about 20,000 cpm	Scaler plus electromagnetic register	Sludge, algae, mud, etc., left after water is filtered off. Samples are dried, and sometimes ashed. Counted on small watchglass. High self-absorption factor
Background—20,000 cpm	"	"
One model has 4,000 and 40,000 dpm full scale. Another model reads up to 5×10^6 dpm	Directly calibrated microammeter	Window may be brought to within $^3/_{16}$ in. from the source. Time constant about 10 seconds
A multitude of models available, reading from 2.5 mrep/hr to about 20,000 mrep/hr full scale	Directly calibrated microammeter	Very useful instrument. Chamber wall has atomic number similar to that of tissue. Reads dosage rate quantitatively. Some are gamma only. Some are air-filled, others are Freon-filled. Some models have chambers attached to a long cable to allow remote monitoring of high intensities
Background to about 20,000 cpm	Audible signal, or count-rate meter. Some use flashing neon bulb to indicate radiation	Should be used strictly as a non-quantitative device. Very useful in tracking down beams, leaks, etc.; then quantitative ionization chamber instrument may be used to measure the dosage rate. Probes usually have an aluminum slide for differentiating between beta and gamma rays
0.1 and 1 rep/hr full scale	Rate-of-drift of quartz fiber in built-in electroscope	Scale is calibrated in terms of time required for fiber to pass between two given marks. Highly reproducible results in low ranges. One model is air wall; can detect alpha's as well. This model uses interval between two neon flashes as a "stopwatch." Other model requires external timing arrangement

Nature of Measurement	*Type of radiation*	*Approximate allowed limits of intensity, dose or contamination*	*Measurement device suggested*
	Slow neutrons	12.5 mrem/hr for 8 hr (1,250–4,700 thermal neutrons/cm²/sec)	Boron trifluoride (gas-filled proportional Counter
	Fast neutrons	12.5 mrem/hr for 8 hr (*see introductory remarks for flux*)	Differential ionization chambers (Chang & Eng)
B. Constant-operation monitors	Beta, Gamma	12.5 mrep/hr for 8 hr	Ionization chamber
1. Dosage-rate indicators			
			GM tube (30 mg/cm² wall)
2. Integrating type (total-dose indicators)	Beta, Gamma	100 mrep/day	Ionization chamber
C. Detection of contaminated surfaces	Alpha	~ 1000 dpm per 100 cm²	Ionization chamber with large surface nylon window
			Air-proportional counter. Large nylon window
	Beta, Gamma	~ 2000 cpm per 100 cm²	GM tube mounted in probe. Either 30 mg/cm² wall or mica end-window type tube used
		About 2 mrep/hr	"Air-wall" ionization chamber
D. Counting smears	Alpha	About 25 dpm	2 in² of Bond paper rubbed over 12 in² of surface. Counted in parallel-plate proportional counter
	Beta, Gamma	About 300 dpm	Same paper and method. Counted under end-window GM counter.

Approximate range of measurement device	*Indicating method*	*Comments*
Practical limit about 20,000 cpm	Headphones or loudspeaker. In some cases a calibrated microammeter is used	May be used in the presence of gammas. Pulse is caused by alpha resulting from B^{10} $(n, \alpha)Li^7$ reaction. May be used for fast neutrons by shielding counter with a sleeve of hydrogenous moderator. This slows down fast neutrons so they may be detected
From about 0.003 to about 300 times tolerance	Rate-of-drift of quartz fiber of Lindemann electrometer	May be used in presence of gamma. Twin chambers mounted on electrometer. One is argon-filled, other butane-filled and polyethylene-lined. Gamma currents balance out. Neutron current produced in butane chamber registers
Various models available, up to about 2,000 mrep/hr	Calibrated microammeter. Alarm may be installed to sound when any preset dosage rate is reached	Usually very stable, trouble-free instruments. A-C operated. May be lined with boron to indicate slow neutron levels
Background—20,000 cpm	Count-rate meter	Not quantitative, but may be used to warn when various preset levels are reached
Various models with different levels available	Electromagnetic register	Meter integrates exposure up to a preset dose, then resets itself, at the same time entering one dose on the register
Zeuto has two scales—4,000 and 40,000 dpm full scale. Zeus reads up to 5×10^6 dpm	Calibrated microammeter	Calibration performed with essentially pure alpha emitters. Time constant about 10 seconds. Air-wall chambers may be used to measure beta-gamma activity also
About 10 dpm per cm^2 to about 20,000 dpm	Audible signal or count-rate meter	A-C operated or portable. Immediate response; very useful for rapid surveying of surfaces. May be calibrated with pure alpha emitters
Background—20,000 cpm	Audible signal or count-rate meter	The end-window type will detect soft betas, and even some alphas
Various models with scales from 2.5 mrep/hr to 20,000 mrep/hr available	Calibrated microammeter	For dosage rate measurements
Background—20,000 cpm	Scaler plus electromagnetic register	Standard smear methods vary. Tolerances for exposed surfaces are lower than those for protected surfaces
Background—20,000 cpm	"	"

Radiochemistry

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RADIOCHEMISTS are concerned mainly with materials which may be emitting alpha particles, beta particles, or gamma rays. As the use of neutrons is one of the easiest manners in which materials may be rendered active, we must also concern ourselves with these. As a result we shall endeavor to present some general methods for the preparation of sources and details of measurement in the estimation of alpha, beta and gamma activities. The production and measurement of neutrons in a laboratory, as distinct from the pile, will also be discussed.

Alpha, beta, gamma activities and neutrons all differ physically, and from the measurement point of view are distinguished chiefly by the manner in which they interact with material, lose energy, and produce ionization. Particular attention must be paid to the understanding and operation of those instruments used in the measurement of radioactivity. The counter is as important to the radiochemist as the analytical balance is to the analyst—and more so.

Two types of instruments are usually used: (1) the counter, which responds to individual pulses, and (2) integrating instruments.

ALPHA PARTICLES

The alpha particle is a He nucleus stripped of its outer electrons. The initial energy is usually of the order of about 4 Mev. The specific ionization (ionization per cm path) is very intense, so that all the ions are found in a very small space. Due to the intense ionization the range is very short, about 4 cm in air, and thus the particle can be stopped by very thin layers of material, *e.g.*, paper.

Preparation of Samples

As a result, samples prepared for alpha counting must consist of very thin layers, to ensure a minimum of self-absorption. Originally the sample may consist of a solution or a precipitate. In either case a flat tray of platinum, gold or stainless steel is used to hold the sample. The solution is evaporated on the tray, usually using an infra-red lamp as a convenient, not too intense, source of heat. It is sometimes convenient to spread a thin ring of lacquer around the edge of the tray, to confine the solution and prevent its spreading over the edges. If the material is in the form of a precipitate, care is taken to ensure uniform spreading, absence of lumps, etc. The mass thickness should be no greater than about 0.1 mg/cm². If the material is crystalline it is centrifuged, washed, slurried with the wash solution, and then spread uniformly in the tray. Except in very special cases, centrifugation is preferred to filtration, because adsorption effects are cut down, and greater speed of operation is obtained. This speed may be extremely important if the activities are short-lived. Unless volatilization of the active material occurs, the tray plus contents should be

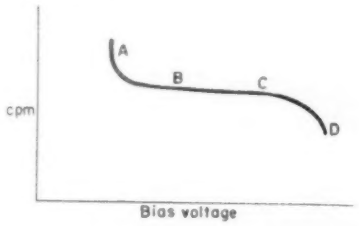
heated to a dull red heat to decompose hydrates which would contribute to absorption of the alpha particles.

The sample is now ready to be counted. Open samples should never be carried into a counting room. The tray is picked up with clean forceps and deposited in a Petri dish. (Care taken to avoid contamination of sample by the instruments or instruments by the sample is very important and cannot be emphasized too strongly.)

Measurement

The sample is placed on the lower plate of a parallel-plate ionization chamber, which consists of two plates separated by about 5–10 mm of air.

Before any actual counting is done, a bias curve for the alpha counting arrangement must be obtained. A reference source of some long-lived alpha-emitting material is made up to give about 3,000 counts per minute. If this is now inserted in the chamber and counts taken at varying bias voltages on the pulse height discriminator, the following type of curve should be obtained:



In part *A* of the curve we will be counting tube noise and spurious counts. The length of the ionization tracks in the chamber will vary due to the geometry of the plate, the sample distribution, etc. In part *B*, the flat part of the curve, which should be about 6 volts long, all the ionization due to long and short tracks is being counted. As the bias voltage is increased, point *C* is reached, which is the part where the shortest tracks are only just being counted. As the voltage is increased the curve at *D* is obtained. We are now cutting out the shorter tracks, counting only the longer ones. For the counting of samples on a comparable basis, the discriminator should be set so that all measurements will be taken at a point in the middle of the plateau. For absolute measurements, a bias curve should be taken with the original sample and the plateau extrapolated back to the ordinate axis to give the true number of counts. This procedure is very important if, as is often the case, the plateau is not flat but has a gentle slope.

Knowledge of the efficiency of the counter is not necessary if only comparative measurements are to be made. However, a determination will have to be made if absolute measurements are being carried out. At first thought, it would seem that the maximum efficiency of the counter can only be 50%, as only those particles which emerge in the direction of the collecting electrode will be counted. However, some compensation due to backscattering occurs, and efficiencies of 55% are not uncommon, if the material of which the tray is composed is a heavy element.

It is very important to avoid contamination of the instrument, which gives spurious counts. In any case, background counts should be taken frequently, i.e., counts with the sample removed and another clean similar tray in position. It is very difficult to obtain materials which are completely inactive. Due to natural contamination there will be about 10^{-4} alphas per cm^2 per minute emitted from a freshly cleaned surface. This will vary by a factor of 10, depending on the choice of material. Lead is very bad in this respect. A natural background of 1–2 counts per minute should be a maximum in an alpha counter.

The number of ions formed by betas

in a given distance will be about $\frac{1}{100}$ that formed by alphas. Thus alphas can normally be counted in the presence of betas. If greater than 10^6 betas are present, superposition occurs and spurious counts may be found. This can often be avoided by the use of a magnetic field in conjunction with the ionization chamber to deflect the beta particles out of the ion chamber.

Corrections

If two experiments are to be compared, or two counts in the same experiment compared, there are some corrections which have to be introduced to make certain that the comparison is an accurate one.

Statistical fluctuations. The emission of the particle or ray by a radioactive substance is a random effect and statistical fluctuations must be considered. If the same sample is counted a large number of times, the average deviation from the true count is equal to the square root of the total number of counts recorded in all the measurements, $D = \pm \sqrt{n}$. If a deviation of no more than 1% is needed, then 10,000 counts must be recorded, so that for a convenient rate of 3,300 counts per minute one must count for three minutes. In this case, $D = \pm \sqrt{10{,}000} = \pm 100$, or $\pm 1\%$ of the number of counts recorded. The probable error, *P.E.*, equals $0.6745D$.

The above should especially be taken into account in work with materials with low counting rates.

Superposition. If the counting rate is very high, the counter may frequently fail to distinguish between consecutive particles. This happens if they occur within a certain time interval known as the resolving time of the instrument. The resolving time should be determined experimentally and corrections made if there is appreciable loss from this cause. This can be ascertained by adding aliquots of a known activity and seeing if the relationship between aliquots added and counts recorded is linear.

Radioactive growth and decay. Corrections may have to be introduced to compare two substances measured at different times. The growth and decay curves can be very complex.

Self-absorption. Even in thin samples self-absorption may not be negligible, and a correction will have to be introduced. This factor may be obtained by use of a standard amount of activity and varying weights of carriers.

Many instruments of the integrating type have been used to measure alpha radiation. The oldest and probably the most reliable of these is the quartz-fiber electroscope. The fiber is charged up to a certain potential and the natural leak measured. Then the fiber is recharged and the source introduced. The time to leak a definite number of divisions is then measured. For comparison the same length of scale is always used, thus keeping the statistical error constant. These instruments are very suitable for laboratory use and usually are reasonably priced or easily constructed, now that high-grade insulators, such as polystyrene, are readily available.

BETA PARTICLES

Beta particles are high-speed electrons with energies ranging from 0.1 Mev to several Mev. They are more penetrating than alpha particles, and will pass through moderate thicknesses, several mg/cm² of material, before being stopped.

Preparation of Samples

The same procedure as for the preparation of alpha samples may be adopted for the betas. The mass thickness may, however, be much greater. The samples may be placed on thin glass slides and covered with cellophane to prevent contamination and loss of

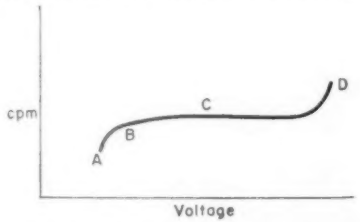
material. Aluminum trays may be used. Here the sample can conveniently be covered by a thin layer of collodion.

Measurement of Radiation

Since beta particles have a fairly high penetration, thin windows can be used on the counters. The Geiger-Müller counter is used very often for the measurement of beta radiation. Chemists usually use an "end-on" type of Geiger.

The Geiger is mounted on a base in a lead box or "castle" with walls two inches thick. This serves to cut down the background intensity due to cosmic radiation and adjacent sources in the laboratory. The base may be slotted to allow samples to be placed on any one of three shelves, thus varying the efficiency of the counter by changing the solid angle it subtends at the source.

In operation of the counter the following technique is usually adopted. A standard "reference" source, giving about 3,000 cpm, is made, usually of uranium in equilibrium with its decay products. This is placed under the counter and the voltage increased until



the counter starts operating. A graph is plotted of the number of counts per minute vs. potential. *A* is the "threshold" where the counter starts operating. *B* is designated as the area of "limited proportionality." As the voltage is increased the Geiger portion *C* is reached, the curve flattens out, the pulses are all of equal height. As the potential is increased the curve starts to rise again in the *D* region. Here the potential is great enough to cause continuous discharge, and breakdowns occur in the counter.

The counter should be operated in the middle of the "plateau" region, so that small fluctuations in voltage will not cause appreciable changes in counting rate. The stabilization of the voltage is much more critical than needed for alpha counting. Even when the above procedure is adopted, fluctuations do occur. It is advisable to "bracket" every count of a sample with two standard readings, and if the variations of the standards are greater than expected due to statistical fluctuations, corrections must be made. The standards are also very valuable if comparisons are to be made between measurements made on different days.

The plateau of a counter should be roughly 150 volts long, showing no greater than a 10% change per 100 volts. As the counter deteriorates, the threshold potential increases, the plateau becomes less flat, and the pressure in the counter increases.

The background of the counter should be about 20 counts per minute, due to cosmic radiation. Contamination of the counter or castle will, of course, increase this.

The geometrical efficiency of the counter varies considerably due to scattering of the betas. It is advisable to cut down this scattering. This can usually be done by lining the lead castle with aluminum and having the sample holder also made of a material of low atomic number, since the scattering is proportional to the square of the atomic number.

Corrections

Statistical fluctuations. Same as for alpha counting.

Superposition. The resolving time of the Geiger plus scaling circuit must be determined. This can easily be done

by irradiation with neutrons of a material with a known decay period, e.g., 54.5-min In. The decay of the In is then measured and should be exponential within the statistical limits. Any departure from this will be due to lack of resolution of the apparatus.

Growth and decay. Same as alpha.

External absorption. The external absorption of the sample must be obtained in order to correct for the absorption in the sample window and air gap between sample and sample window. This is obtained by interposing Al absorbers between the sample and the window. The absorption is roughly exponential, with a tail, if any is present, due to gamma radiation. In this manner the amount of material required to cut the radiation down to half of its intensity, $\epsilon_{1/2}$, is found. The value $\epsilon_{1/2}$ is characteristic of the beta emitter and may serve as a source of identification. The range of the beta ray is also characteristic of the emitter.

Self-absorption. Unless very high intensity sources are used, the sample which is to be checked for activity will have a finite thickness due to added carriers or the fact that it is inseparable from the starting material. For comparison purposes, corrections must be made for the amount of radiation absorbed by the material itself, the self-weakening half-thickness $\eta_{1/2}$. The correction can sometimes be avoided by use of either of the following methods.

1. For comparable experiments the same mass of material may be used each time. This is usually very inconvenient, especially where short-lived emitters are concerned.

2. The amounts of material used may be so thick that only radiation from the top layers will reach the counter, *i.e.*, a mass thickness greater than the range. This is usually not very practical, since the sample intensity will be cut.

$\eta_{1/2}$ may usually be obtained as follows: The active material is precipitated and varying amounts are placed in trays and the activities measured. The measured activity per unit weight of sample in each case is then calculated (apparent specific activity), and plotted against the actual mass of the sample which was counted. On account of the exponential absorption of beta rays in matter, one finds a logarithmic relation between the mass of the sample and the apparent specific activity for small values of the mass. If there were no self-weakening, the apparent specific activities would all be the same, but as self-weakening is always present, the curve shown above is obtained. By extrapolation back to zero mass, the true specific activity may be obtained. $\eta_{1/2}$ may also be read off the graph.

If the activity has an appreciably short half-life, decay curves are obtained for the various mass thicknesses and the above procedure used to determine the curve at a definite time.

As a rough guide, the relation $\eta_{1/2}/\epsilon_{1/2} = 2.2$ may be used. This has been determined experimentally for a large number of radiations, and found to hold roughly true within limits. Care must be taken in the determination of $\eta_{1/2}$ and $\epsilon_{1/2}$ where the beta radiations are complex, as errors may easily be introduced.

Backscattering. If the results to be obtained are only relative comparisons between different activities of the same β-emitter, no correction need be introduced for the amount of radiation backscattered into the counter by the sample holder or material surrounding the counter. If a knowledge of the number of disentegrations per minute is desired or if different β-emitters are being compared, a correction factor is necessary. This can be obtained in two ways.

1. Experimental curves are obtained for increase in counts/minute with increasing mass of the backing material for β-emitters of various energies.

2. All measurements are made with sufficient backing material present to give "saturation" backscattering. The correction factors are then obtained from experimentally determined curves.

This last method is preferred since small changes in tray thickness do not affect the result.

The usual window thickness in a Geiger is of the order of 5 mg/cm². Unfortunately the betas of many of the radioactive isotopes have energies so small that even such a thin window will cut out a great deal of the radiation, e.g., C^{14} (maximum beta energy 154 kev). For such isotopes special techniques have to be evolved. Special thin-window counters can be made with windows about 2 mg/cm². These are usually very fragile. The windowless counter technique is sometimes used. A very simple windowless counter may be constructed using a desiccator and simply removing the window from an ordinary G-M counter. The entire desiccator is filled with the gaseous mixture. Absorbers can be swung into position by means of an externally controlled magnet. An external standard may also be used.

Sometimes the material to be measured can be introduced directly into the counter in the form of a gas, e.g., CO_2. This is a very efficient method, giving a geometry of almost 100%.

Other special techniques include integrating instruments, such as electroscopes and ionization chambers. In general, the electroscope will be $\frac{1}{50}$ to $\frac{1}{500}$ as sensitive as the Geiger, while the ionization chamber with an FP-54 electrometer tube will be $\frac{1}{10}$ to $\frac{1}{100}$ as sensitive as the Geiger.

Where directional effects are required, two Geigers may be connected so that only counts which register in both Geigers will be recorded. This is known as "coincidence" counting and is a potent technique in cosmic ray work, standardization of sources, etc.

GAMMA RAYS

Gamma rays are high-energy photons of very short wavelength. Energies run from the X-ray region to several Mev. They are very penetrating. Gamma rays do not lose energy gradually as do alphas and betas, but ionize by collision with one or a very few electrons. The electrons further ionize by collision as previously described, and thus the gammas are detected. Gamma rays interact with matter in the following three ways: photoelectric absorption—the photon transfers all of its energy to an electron and ejects it from the atom, usually from *K* or *L* shell; Compton scattering—the primary photon ejects an electron by transferring to it only part of its energy while the remainder appears as a scattered photon; pair-production—the energy of the photon is converted into the mass and kinetic energy of a positron-electron pair. This is a reaction between the photon and the nucleus, and usually occurs only with high-energy gammas.

The ordinary Geiger is normally about $\frac{1}{100}$ as efficient for the gammas as it is for the betas. A layer of heavy element, e.g., lead, is placed in front of the window to act as a "radiator" and the betas are measured by the Geiger. It is advisable to place a layer of aluminum under the lead to cut out any betas originally present in the sample. If this is not done the betas will cause the generation of Bremsstrahlung, or continuous X-radiation, which would be absorbed in the lead, giving rise to electrons and thus an increased effect. This conversion of the kinetic energy of the electron to electromagnetic radiation is proportional to the square of the atomic number of the stopping elements.

NEUTRONS

The neutron is a particle of mass 1 and no charge. Because of this lack

(Continued on page 141)

SOME SUGGESTED USES OF ISOTOPES

THE PURPOSE of this section is to list possible uses of radioisotopes in industry. The ideas presented below have been collected from bulletins, reports and scientific journals and no accuracy, originality or practicability is claimed for them. This material is taken from a report (CRIB-399) by G. H. Guest, National Research Council of Canada. It was prepared for use at a two-day conference on industrial uses of radioisotopes, held in December, 1948, in Ottawa. An asterisk indicates that some work has been done on the item marked.

*1. Calibration of ultra microbalances with alpha-active material.** The calibration of microbalances can be improved by using substances with high alpha activity since the activity can be determined more accurately than the weight.

2. Use of tracers in electroplating baths to determine concentration of an ion. The depletion of minor constituents in electroplating baths might be followed with a radioisotope.

*3. Use of tracers in the analysis of a product from a reaction mixture of known composition.** Analysis of a product for impurities may be made by the addition of a radioisotope to the reaction mixture in a definite proportion to the weight of the element or elements sought. After separation of the product from the reaction mixture it is counted for the element sought (which would be an impurity or a principal constituent). The supernatant would be "counted" for an element of the product to determine the extent of the reaction.

4. Use of tracers to study the cause and cure of foaming and priming. Tracers can be used to study the cause and cure of industrial foaming and priming, the effect of different dissolved salts and antifoams.

*5. Distribution measurements of textile finishes.** By the use of tracers, studies have been made of the distribution of lubricant on rayon fiber in order to evaluate the role of the lubricant in subsequent dying and spinning operations.

6. Flow studies of components of process operations. Modifiers in process operations, such as plastic manufacture, might be followed, giving information on mechanism and value of such materials.

7. Process rate studies. Rate studies on industrial processes can be of value in understanding of the processes. Tracers have wide application in this field.

8. Product control. Any reaction involving inorganic materials where freedom from impurities is important. Examples are: analytical reagent manufacture, pigment manufacture, ceramic manufacture.

*9. Use of tracers in distillation.** Determination of equilibrium data and measurement of operating efficiency are paramount problems; only poor analytical methods exist in many cases. Radioactive tracers may be used to obtain binary equilibrium data and vapor-liquid equilibrium data of pre-

cision in dilute ranges. In the determination of the number of plates in high-efficiency fractionating columns it is often difficult to analyze for purity at the top and bottom of the column. The addition of the suitable radioactive tracer into the feed would make this analytical problem relatively simple. Such a method might be particularly useful in the separation of the C-4 hydrocarbons for synthetic rubber and the purification of rare gases such as xenon, argon, and neon from air. In the distillation of materials to high purity where concentrations are not sufficient for automatic control by temperature, the concentration of tracer-containing impurities in the charge may be used for control and continuous indication of distillation progress. If numerous samples are to be withdrawn, the tracer technique offers an easy method of analysis, particularly in multi-component systems. In experimental studies of batch distillation, radioactivity can provide an easy means of following the movement of low boiling components up through the fractionation column, and the progressive concentration of these components at various points as equilibrium is approached.

10. Use of radioisotopes in metering fluid. Radioisotopes can serve as metering fluid in the usual method of dilution measurements and have a decided advantage in case of analysis.

11. Use of tracers to determine the effectiveness of scrubbing gases. In some cases it is desirable to scrub materials from gases so that they are present in extremely small quantities in the exit gas. Radioactive tracers of these materials may simplify the analytical problem. Often it is difficult to analyze the efficiency of absorption columns because of the difficulty of analyzing the excess gas phase. A radioactive tracer might afford a simple and easy method for overcoming this problem. Absorption data in very dilute ranges can be obtained using a radioactive tracer.

*12. Use of tracers in the analysis of equilibrium data and the relative efficiency of various absorbents.** Practical application has already been made in the case of the absorption of xenon by silica gel where the 5.4-day active xenon was successfully used as a tracer.

*13. Use of tracers to determine separation efficiencies in centrifugation, filtration, etc.** Tracer procedures have proven to be of great value in detecting the presence of finely divided solids in liquids which have passed through a separation process. Thus, an easy means of calculating separation efficiencies for centrifugation, filtration, etc., is provided.

14. Determination of the homogeneity of a mixture after various times and degrees of agitation. The degree of homogeneity of a mixture, after various times and degrees of agitation, can be determined by the withdrawal of samples at several points and the comparison of the composition with the proportion of components entering the vessel. The tracer technique is readily adaptable to the study of solid in liquid systems and suspensions of solids in liquids.

15. Determination of the settling rate of a suspension. The rate of settling of a precipitate in a closed tank can be followed with ion chambers or counters if a tracer is carried down with the precipitate which is settling.

16. Use of tracers in process control to determine concentration of an ion in continuous operation: Middle processing baths—cleaning, fire-growing, coating; electroplating baths—minor constituents; rayon processing baths—minor constituents; textile processing baths; pulp and paper manufacture; and gas-reacting system.

17. Measurement of linear velocity of liquid flow with tracers. The linear velocity of liquid flow in a pipe can be

measured by timing the successive appearance of activity at two points along its length. The method is of special value where circumstances are such that sampling is impossible; the activity can be measured through the pipe walls.

18. Use of tracer to determine the effect of tracer impurities upon the physical properties of synthetic fiber. It is known that tracer amounts of manganese are deleterious to rayon. Studies to establish the magnitude of the effect as related to the amount of impurity could be furthered by the use of a tracer in making analysis, since the colorimetric analyses are not sensitive enough under the conditions of experimentation.

19. Use of radiocarbon to study co-polymerization. In the relatively new field of co-polymerization in which polymers are produced from two materials neither, or only one, of which will polymerize by itself, the problem often arises as to the composition of such a polymer and also whether it is a real co-polymer or a component polymer with the other component imbedded in it.

*20. Use of tracers for determining vapor pressures.**

21. Use of tracers for rapid discovery and isolation of catalyst poison.

*22. Use of radioisotopes in studying adsorption processes.** Tracers may be used for showing the distribution of material throughout a resin bed, and for the detection of break-through.

*23. Use of activity to stabilize balance performance.** To minimize the influence of static charges upon the accuracy of weighing, a radioactive source is placed in the balance chamber. This application has been used for many years and is not unique.

24. Development of "self-promoting catalysts." It might be possible to develop catalysts in which the presence of radioactive material causes repeated renewal and extension of the active surface.

25. Influence of radiation on interpolymerization of organic compounds. Certain systems cannot be made to interpolymerize although at least one of these components is readily polymerizable. Examples are: propylene plus styrene, isobutylene plus styrene and isobutylene plus butadiene.

26. Cracking of petroleum oils with intense radiation. Low-temperature cracking of petroleum may be possible.

27. Use of radiation for nitrogen fixation.

28. Hardening of metals or plastics with intense radiation may be possible.

29. Migration of metallic ions into insulating compounds.

30. Mechanism of adsorption and settling rates of additions in oil corrosion.

31. Friction and wear tests—gasoline engine.

32. Mining. Mechanism of flotation of minerals—heterogeneous adsorption at low concentrations—cationic collecting agents; oil, gas, and water permeability in three phase flow in porous media; ion exchange equilibria—well logging; and injection water flow profiles—petroleum recovery.

33. Quality control. Thickness of silicone film on rubber—adhesion; height gage—cupola level; and thickness gage—metallic and plastic sheets.

34. Mechanism of hardening of evaporated mirrors by strong oxidants.

35. Efficiency of mixing graphite and liquids.

36. Determination of tarnish resistance of silver plate.

37. Determination of transfer of printing ink from printing plate to paper.

PROPERTIES OF MATERIALS

Photographic Recording Media

HERMAN YAGODA
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National Institutes of Health
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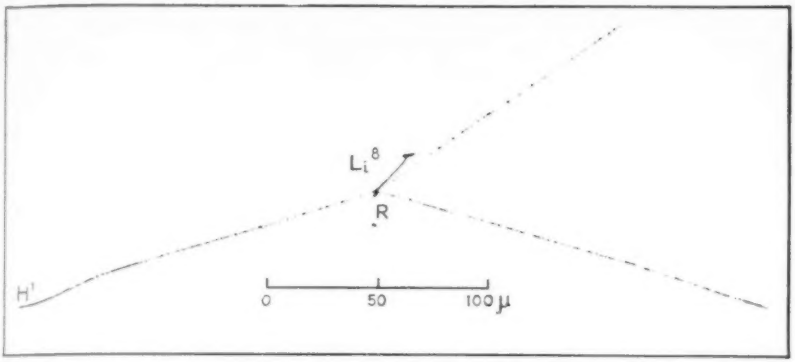
Alpha Particles

Objective	*Emulsion*	*Remarks*
Autoradiographs or alpha-ray patterns	Eastman NTA, 25 microns, or Ilford C_2, 50 microns thick	Provide a selective alpha-ray pattern of good contrast and permit microscopic resolution of structures of about 20 microns diameter. About 10^7 alpha particles per cm^2 are necessary for macro observation of detail (*1, 2*)
	Eastman lantern slides	Show good detail at 1 to 10 × magnification. Also beta sensitive. Exposure about 10^6 alpha particles per cm^2
	Eastman No-screen X-ray film	Fast, but detail is poor. Beta- and gamma-ray sensitive
Alpha track counting (2 to 10 Mev)	Eastman NTA or Ilford C_2 coated 50 microns	Individual alpha particle tracks observed at magnifications above 200 ×. Moderate background fog of beta and gamma radiations does not interfere with alpha track visibility. Alpha tracks readily differentiated from fast proton ($\geq$ 4 Mev) trajectories by variation in developed grain density
	Eastman NTB or Ilford B_2, coated 50 microns	Record exceptionally robust alpha tracks, permitting counting at low magnifications. Record tracks of slow electrons; fogged by excessive doses of beta particles
Trajectories of fast alpha particles	Eastman NTA or NTB coated 100 microns, Ilford C_2 coated 200 microns	Will record alpha particles with energies of about 400 Mev. Thick emulsions increase probability of recording complete range, but require special processing (*3*); see figure on next page.

Protons

Scattering	Ilford C_2 coated 50 microns	In exposures under high vacua the emulsion tends to peel. May be avoided by painting edges with shellac



Nuclear evaporation recorded at Echo Lake, Colorado, altitude 3,200 meters, showing ejection of a proton H^1, a Li^8 fragment decaying into two alpha particles, a heavy short recoil track *R* and the tracks of two other unidentified particles which leave the emulsion layer.

PLATE: Ilford C_2 coated 200 microns thick.

DEVELOPER COMPOSITION: Hydroquinone 2.0 g, Na_2SO_3 20 g, KBr 1.0 g, Na_2CO_3 10 g per liter of distilled water.

DEVELOPMENT: Chill developer solution and glass tray to 10° C. Insert plate in developer and place in a refrigerator at 5° C for 1 hour. Transfer tray to water bath at 30 to 35° C. Dilute developer with equal volume of water at 20–22° C, adding water slowly and with adequate agitation for uniform mixing. When temperature reaches 22° C, allow development to continue without further agitation for 30 minutes at 22 to 20° C.

FIXATION: Siphon off developer and fix in 30% sodium thiosulfate until clear (about 2–3 hours, depending on rate of agitation and temperature). When plate is nearly clear, complete fixation and harden by 30 minute immersion in Eastman F5.

RESIDUAL PROCESSING: Wash in cold filtered tap water for 2 to 3 hours. Soak for 10 minutes in an aqueous solution containing 1% glycerine and 0.1% Aerosol wetting agent. Drain thoroughly with aid of filter paper and dry plate in horizontal position. Coat borders of plate with shellac, shortly after drying, to prevent curling of gelatin edges under conditions of low humidity.

This procedure gives uniform depth development, and the background fog grains are reduced in size. Although the minute fog grains are evident microscopically they do not reproduce on high contrast photomicrographic media. The tracks of densely ionizing particles are readily evident at 200 ×, and, at higher magnification, the tracks of fast protons and σ-mesons are also visible. The method facilitates track discrimination by grain counting.

Protons (Continued)

Objective	*Emulsion*	*Remarks*
Fast proton recording	Eastman NTA or NTB coated 100 microns, or Ilford C_2 coated 200 microns	Record tracks of particles with energies below 50 Mev
Particle identification	Ilford C_2 coated 100 microns	Slight underdevelopment facilitates discrimination of tracks produced by particles differing in ionizing power, as described in figure

Objective	*Emulsion*	*Remarks*
Deuterons		
Fast deuteron recording	Ilford C_2 coated 200 microns	Tracks of particles with energies below 100 Mev record
Tritons		
Track recording	Ilford C_2 coated 200 microns	Upper limit about 150 Mev
Particle identification	*ibid.*	Differentiation between triton and alpha particle tracks facilitated by underdevelopment, or differential fading effects (*4, 5*)
Meson Particles		
Identification	Ilford C_2 coated 100 or 200 microns	Tracks of slow mesons of about 300 m_e with energies below 8 Mev are readily differentiated from protons by grain density and degree of scatter (*6, 7*)
Fission Fragments		
Selective registration in presence of predominating alpha-particle flux	Eastman NTC coated 25 microns or Ilford D_1 coated 50 microns	Details described in (*8, 9*)
Range measurements	Ilford C_2 desensitized with uranyl acetate	Details described in (*10*)
Fission pairs accompanied by long range alpha particles	Eastman "fine-grain alpha" loaded from saturated aqueous uranyl acetate (*11*). Ilford C_2 coated 100 microns loaded from 0.5 to 1.0% uranyl acetate in 10% acetic acid (*10*)	Alpha particles from uranium differentiated from fission fragments by means of grain density. Discrimination enhanced by underdevelopment. The loading process weakens sensitivity so that protons arising from neutron recoils do not record tracks
Heavy Charged Particles		
Recording and identification	Eastman NTA 100 microns, or Ilford C_2 coated 200 microns	Differentiated from other particles by range and grain density considerations. Particles of about 10 Bev also exhibit short delta rays along the trajectory (*12, 13*)
Fast Neutrons		
Identification as proton recoils	Eastman NTA or NTB emulsion coated 100 microns or Ilford C_2 coated 200 microns	Collisions between fast neutrons and hydrogen nuclei are recorded by the track of the proton recoil

Objective	*Emulsion*	*Remarks*
Fast Neutrons (Continued)		
Identification of stars from $B^{10} + n^1 \rightarrow 2He^4 + H^3$ reaction	Ilford C_2 plates boron-loaded and coated 100 to 200 microns thick	The more compact tracks of the alpha and triton particles permit evaluation of neutron energies below 100 Mev from range and momentum considerations (*14*)
Identification of stars from $B^{11} + n^1 \rightarrow Li^8 + He^4$	Ilford C_2 plates boron-loaded and coated 200 microns	These events are readily recognized by the "hammer track" produced by the decay of the Li^8-fragment (*15*)
Identification of stars from $C^{12} + n^1 \rightarrow Li^8 + He^4 + H^1$	Eastman NTA or Ilford C_2 coated 100 to 200 microns	Observed in plates exposed to cosmic radiation (*16*)
Slow Neutrons		
High neutron flux measurements	Eastman NTA or Ilford C_2 plates	Short-range proton tracks result from slow neutron capture of the gelatin by N^{14} (*17*). Cross section of $N^{14}(n^1,H^1)C^{14}$ reaction is low
Low neutron flux measurements	Ilford C_2 plates boron-loaded	Short, dense alpha tracks originate from the $B^{10}(n^1,He^4)Li^7$ reaction (*3*)
Slow neutron identification	Ilford C_2 plates lithium-loaded	The characteristic tracks originating from the $Li^6(n^1,H^3)He^4$ reaction provide a selective indicator for presence of slow neutrons (*3*)
Low neutron flux	Eastman NTC 25 microns thick in contact with foils of U^{235}	Fission tracks serve as index for neutron flux (*3*)
Beta Particles		
Localization of radioisotopes in tissue blocks	Eastman "lantern slides"	Good definition of macro segregates (*3*)
	Ilford line film	Effects resolution of 10 to 15 microns. Low speed, necessitates very active tissue (*19*)
	Eastman "no-screen" X-ray film	Fast, suitable for autoradiography of long-lived isotopes (*19*)
Permanent tissue emulsion autoradiographic patterns	Eastman NTB stripping film	Details described in (*20*, *3*)
	Eastman type M stripping film, or Ansco Reprolith ortho stripping film	Details described in (*21*)
	Ilford halftone stripping emulsion	Achieves a resolution of 3 to 5 microns. Slow, requires an exposure about 4 times that for fast X-ray film (*18*)
Beta-ray intensity	Eastman "no-screen" X-ray film	Fast, suitable for weak sources

Beta Particles (Continued)		
Objective	*Emulsion*	*Remarks*
	Eastman "contrast lantern slide"	Exhibits linear density response over wide range of intensity
	Ilford type B_2	Low background fog, faster than lantern slides
	Eastman NTB	Low background fog. May record individual tracks of soft beta particles
Track registration	Kodak Ltd. NT2a or, Eastman NTB emulsions	Record short highly scattered tracks of slow electrons and soft beta particles such as originate in the decay of RaD, C^{14} and Rb^{87}(*22*)
Gamma Radiation		
Dosage measurement	Eastman type K X-ray film	Useful in estimating accumulated doses of 0.05 to 2.0 roentgens
	Eastman type A X-ray film	Covers range of 1.0 to 10 roentgens
	Eastman Kodalith	Covers range of 70 to 700 roentgens
	Eastman type 548-0	Useful for very intense radiation above 2000 r
	Defender Adlux film	Covers a wide range between 50 to 15,000 roentgens (*23*)
Energy evaluation by photodisintegration processes	Ilford C_2 loaded with $Ca(NO_3)_2{\cdot}D_2O$	Energy of photon evaluated from range of protons originating from disintegration of deuterium (*24*)
	Ilford C_2 loaded with heavy water and exposed while wet	Provides high concentration of deuterium nuclei, but proton tracks are of lower grain density (*25*)
	Ilford C_2 loaded with beryllium	Energy evaluated from alpha tracks produced by $Be^9 + h\nu \rightarrow Be^8 + n^1$
	Ilford C_2 coated 100 to 200 microns	Photons with energies of 17 Mev and higher disintegrate C^{12} nuclei producing stars composed of three alpha particle tracks (*26, 3*)

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Counter Fillings

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Simple Gases

Argon. Medium breakdown voltage, high specific ionization

Helium. Low breakdown voltage, low specific ionization

Neon. Medium breakdown voltage, high specific ionization

Air. High breakdown voltage, forms negative ions by electron attachment to the O_2, hence has long resolving time

Nitrogen. High breakdown voltage, high specific ionization

Hydrogen. High breakdown voltage, low specific ionization

Carbon dioxide. High breakdown voltage, forms negative ions on collision between its positive ions and the cathode

Advantageous Mixtures of Simple Gases

Noble gases and mercury. Noble gases have troublesome metastable states which are removed by ionizing mercury atoms; this decreases the

Typical Simple Gases Used in Counters

Gas	*Formula*	*1st Excitation Potential (volts)*	*Ionization Potential (volts)*	*Electron Attachment Probability*	*Negative Ion Formed at Wall*	*Positive Ion From Which Formed*	*Negative Ion Formation Probability*	*Metastable Level (volts)*	*Emission Wavelength (A.U.)*
Argon	A	11.6	15.7	0	—	—	—	11.5	1048
Helium	He	20.5	24.5	0	—	—	—	19.7	854
Krypton	Kr	9.9	13.9	0	—	—	—	9.87	1236
Mercury	Hg	—	10.4	—	Hg^-	Hg^+	10^{-3}	5.43	1850
Neon	Ne	16.6	21.5	0	—	—	—	16.5	736
Xenon	Xe	8.3	12.1	0	—	—	—	8.27	1470
Hydrogen	H_2	11.5	15.4	0	H^-	H_2^+	10^{-5}	—	—
Atomic Hydrogen	H	10.2	13.5	0	—	—	—	None	1215
Nitrogen	N_2	6.1	15.5	0	—	—	—	6.27	—
Atomic Nitrogen	N	—	14.5	—	—	—	—	2.37	1135
Oxygen	O_2	6	12.5	10^{-4}	O^-	O_2^+	10^{-3}	5, 3,	—
					O_2^-	O_2^+	10^{-5}	and 9	
Atomic Oxygen	O	—	13.6	10^{-3}	—	—	—	2, 4, and 9	1302
Carbon Dioxide	CO_2	—	14.4	0	O_2^-	CO_2^+	10^{-4}	5.6	—
					CO_2^-	CO_2^+	10^{-4}		

spurious counts from the noble gases

Neon and argon. Removing the metastable states in neon by small admixtures of argon greatly reduces the breakdown voltage, so that this mixture has thresholds of a few hundred volts

Carbon dioxide and carbon disulfide. The CS_2 exchanges charge with the CO_2 positive ion to prevent CO_2 negative ions being formed as mentioned before

Typical Polyatomic Gases Used in Counters

Gas	*Formula*	*Ionization Potential (volts)*	*Electron Attachment Probability*	*Negative Ion Formed in Gas*	*Energy to Form Negative Ion (volts)*	*Absorption Wavelengths (A.U.)*	*Type of Spectra*
Bromine	Br_2	12.8	10^{-3}	Br^-		1500	Continuous
Chlorine	Cl_2	13.2	10^{-3}	Cl^-		1500	Continuous
Carbon Monoxide	CO	14.1		O^-	20.9		
Nitric Oxide	NO	9.5	5×10^{-5}	O^-	7.0		
Carbon Dioxide	CO_2	14.4	0			1360–600	Bands
Carbon Disulfide	CS_2	10.4				3800–1200	Bands
Hydrogen Sulfide	H_2S	10.4	0	HS^-	3.7	1600–1190	Bands
	HS		10^{-3}				
Nitrogen Dioxide	NO_2	11.0				5700–2200	Bands
Nitrous Oxide	N_2O	12.9	0	O^-	1.7	3000–1760	Continuous
						1520–1056	Bands
						< 1000	Continuous
Sulfur Dioxide	SO_2	13.1	10^{-4}	SO^-	5.7	3800–1529	Bands
Water	H_2O	13.0	0	OH^-	5.4	1240–983	Bands
				O^-	7.5		
	OH	13.8	10^{-4}				
Ammonia	NH_3	10.5	0	NH_2^-	6.0	1620–1450	Bands
				NH^-	3	< 1200	Continuous
	NH^-		10^{-4}				
Acetylene	C_2H_2	11.6				2400–2090	Bands
Carbon Tetrachloride	CCl_4					4600–2300	Bands
Methane	CH_4	14.4				< 1450	Continuous
Chloroform	$CHCl_3$					< 2200	Continuous
Methyl Iodide	CH_3I	10.12				3600–2110	Continuous
						2100–1215	Diffuse
Ethyl Bromide	C_2H_5Br	10.24				2850–1900	Continuous
						< 1700	Diffuse
Ethyl Chloride	C_2H_5Cl					< 1700	Continuous
Pyridine	C_5H_5N	9.8				< 2500	Continuous
Ethyl Alcohol	C_2H_5OH	11.3				1633–1602	Bands
						1518	Diffuse
						< 700	Continuous
Acetone	CH_3COCH_3	10.1				3300–2940	Bands

Quenching Gases and Vapors

Bromine and chlorine. Good quenching agents in low concentration, but give appreciable negative ions, and hence spurious counts, above a few tenths of one percent concentration. Have unlimited life

Methane. Readily available for flow counters. Has short life when used in sealed-off tubes

Ammonia. Decay products show some electron attachment

Acetylene, carbon tetrachloride and similar relatively simple molecules show short quenching life

Ethyl alcohol is the most common quenching agent. Can show marked effect with temperature and is useless below 40° F

Ethyl ether has a high enough vapor pressure at room temperature to make it relatively free of temperature effects

Ethylene is a gas, not a vapor, and is a good quenching agent showing no temperature effect. It also does not react with organic film windows used in beta-ray counters

Tetra methyl lead is a satisfactory quenching vapor but may cause counters to be photosensitive to visible light

Butane when frozen onto the cathode with liquid air shows, thereafter, great photosensitivity

Other good quenching agents without special gas discharge properties include methylene bromide, cyclopropane, pyridine and ethyl bromide

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of charge it is repelled neither by the nucleus nor the external electrons and can be very conveniently used to activate materials.

A very good source of neutrons is, of course, the chain-reacting pile. However, a laboratory can be supplied with a neutron source that is much simpler in construction, and much cheaper.

$$_4Be^9 + {}_2He^4 \rightarrow {}_6C^{12} + {}_0n^1$$

The alpha particles from Ra, Po or Rn may be used. The source is made up by mixing intimately the alpha emitter and the Be. This is necessary because of the short range of the alpha particles.

$$_4Be^9 + h\nu \rightarrow {}_4Be^8 + {}_0n^1$$

$h\nu = 1.627$ Mev or greater

The Be is usually a block outside the source.

Measurement

Slow neutrons.

$$_5B^{10} + {}_0n^1 \rightarrow {}_3Li^7 + {}_2\alpha^4$$

and the alphas are measured as previously described. Thermal neutrons, i.e., neutrons with energy about $\frac{1}{40}$ ev, are measured in this fashion. A convenient source of B is found in BF_3, which is a gas at room temperature and ordinary pressures, and may therefore be used as a counter filling. Metallic boron coatings on the insides of the counter wall may also be used.

A very convenient method of measuring slow neutron intensity is by the use of foils. For example two sources may be compared by irradiation of calibrated in foils in the same geometry and comparison of the beta activities.

Fast neutrons. These are usually measured by high-pressure H-filled ionization chambers. The neutron strikes a hydrogen nucleus, recoil of the nucleus occurs, breaking the binding between the nucleus and the extranuclear electron, and the ionization by the electrons is measured.

Bibliography on Radiation Detection

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THIS BIBLIOGRAPHY was compiled, under the auspices of the Committee on Nuclear Science of the National Research Council, in response to a request from the Nuclear Studies Committee of the Institute of Radio Engineers. It is a selection of some of the more significant references. For more complete bibliographies, the reader is referred to "An International Bibliography on Atomic Energy" (United Nations, Lake Success, N. Y.), Vol. II—Scientific Aspects, Part I-D—Detection of Nuclear Radiation; and the bibliography by Monica Healea [NUCLEONICS **1,** No. 4, 68 (1947), **2,** No. 2, 63 and **2,** No. 3, 66 (1948)].

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BUYERS' GUIDE

*MANUFACTURERS CLASSIFIED BY PRODUCT**

Accessories (*Shields, lead bricks, etc.*)
- Atomic Instrument
- Bar-Ray
- Canadian Marconi
- Flader
- Nuclear Instrument & Chemical
- Radiation Counter
- Technical Associates
- Tracerlab

Counting-rate Meters
- Anton
- Atomic Instrument
- El-Tronics
- General Radio
- Measurement Engineering
- Radiation Counter
- Radioactive Products
- Technical Associates

Electrometers
- Applied Physics
- Cambridge
- Raytheon
- Victoreen
- Well Surveys

Electroscopes (*Lauritsen*)
- Henson

Geiger Tubes
- Able Scientific
- Amperex
- Anton
- Atomic Instrument
- Ballantine
- Canadian Marconi
- Cyclotron Specialties
- Distillation Products
- El-Tronics
- General Radio
- Geophysical
- Herbach & Rademan
- Kelley-Koett
- Martin
- North American Philips
- Nuclear Development
- Nuclear Instrument & Chemical
- Precision Radiation
- Radiation Counter
- Raytheon
- Sylvania
- Technical Associates
- Tracerlab
- Victoreen
- N. Wood

Ionization Chambers
- Applied Physics
- National Technical
- Victoreen
- Well Surveys

Monitors
- Cambridge
- Electronic Associates
- Geophysical
- Kelley-Koett
- Measurement Engineering
- Radiation Counter
- Telemark
- Tracerlab
- Victoreen

Parallel-plate Counters
- Abrams
- Nuclear Instrument & Chemical

Personnel Meters
- Atomic Instrument
- Cambridge
- Kelley-Koett
- Nuclear Instrument & Chemical
- Victoreen

Proportional Counters
- Atomic Instrument
- General Electric
- Nuclear Instrument & Chemical
- Radiation Counter
- N. Wood

Scalers
- Anton
- Atomic Instrument
- Berkeley Scientific
- Canadian Marconi
- Cyclotron Specialties
- El-Tronics
- Flader
- General Electric
- Kelley-Koett
- Nuclear Instrument & Chemical
- Technical Associates
- Tracerlab
- Victoreen

Survey Meters
- Bay Instrument
- Canadian Marconi
- El-Tronics

* Complete names and addresses of companies will be found on page 152.

Survey Meters *(continued)*

Geophysical
National Technical
North American Philips
Nuclear Instrument & Chemical
Nucleonic Corp. of America
Omaha Scientific
Precision Radiation
Radiation Counter
Sylvania
Technical Associates
Tracerlab
Victoreen

Index of Manufacturers

Able Scientific Glass Apparatus Co., 2350 South Cottage Grove, Chicago 16, Ill.

Abrams Instrument Corp., 600–620 East Schiawassee St., Lansing, Mich.

Amperex Co., 25 Washington St., Brooklyn, N. Y.

Anton Electronic Laboratories, Inc., 1226–1238 Flushing Ave., Brooklyn 6, N. Y.

Applied Physics Corp., 30 West Green St., Pasadena 2, Calif.

Atomic Instrument Co., 160 Charles St., Boston, Mass.

Ballantine Laboratories, Boonton, N. J.

Bar-Ray Products, Inc., 209 25th St., Brooklyn 32, N. Y.

Bay Instruments & Development Co., 2200 Woolsey St., Berkeley 5, Calif.

Berkeley Scientific Co., Sixth and Nevin Ave., Richmond, Calif.

Cambridge Instrument Co., 72 East 45th St., New York, N. Y.

Canadian Marconi Co., Montreal, Quebec, Canada.

Cyclotron Specialties Co., Moraga 8, Calif.

Distillation Products, Inc., 755 Ridge Road West, Rochester 13, N. Y.

Electronic Associates, Yonge Street, Toronto, Ontario, Canada

El-Tronics, Inc., 1920 Lincoln-Liberty Building, Philadelphia 7, Pa.

Fredric Flader, Inc., Engineering Physics Division, N. Tonawanda, N. Y.

General Electric Co., Electronics Department, Syracuse, N. Y.

General Radio Co., 275 Massachusetts Ave., Cambridge 39, Mass.

Geophysical Instrument Co., Key Boulevard and Nash St., Arlington, Va.

Fred C. Henson Co., 3511 E. Colorado St., Pasadena, Calif.

Herbach and Rademan Co., 522 Market St., Philadelphia, Pa.

Kelley-Koett Manufacturing Co., 212 West 4th St., Covington, Ky.

H. S. Martin Co., 1916–1920 Greenleaf St., Evanston, Ill.

Measurement Engineering, Arnprior, Ontario, Canada

National Technical Laboratories, 820 Mission St., South Pasadena, Calif.

North American Philips Co., Inc., 100 East 42nd St., New York 17, N. Y.

Nuclear Development Laboratory, P. O. Box 7601, Kansas City, Mo.

Nuclear Instrument & Chemical Corp., 223 West Erie St., Chicago 10, Ill.

Nucleonic Corp. of America, 497 Union St., Brooklyn, N. Y.

Omaha Scientific Supply Corp., 3623 Lake St., Omaha 4, Neb.

Precision Radiation Instruments, Inc., 1101 North Paulina St., Chicago 22, Ill.

Radiation Counter Laboratories, 1844 W. 21st St., Chicago, Ill.

Radioactive Products, Inc., 3201 East Woodbridge St., Detroit 7, Mich.

Raytheon Manufacturing Co., 55 Chapel St., Newton 58, Mass.

Sylvania Electric Products, Inc., 70 Forsyth St., Boston 15, Mass.

Technical Associates, 3730 San Fernando Rd., Glendale 4, Calif.

Telemark, Inc., 79 Prospect St., Stamford, Conn.

Tracerlab, Inc., 55 Oliver St., Boston, Mass.

Victoreen Instrument Co., 3800 Perkins Ave., Cleveland, O.

Well Surveys, Inc., 624 East 5th St., Tulsa 3, Okla.

N. Wood Counter Laboratory, 5646 S. Harper St., Chicago, Ill.